

GLOBAL OZONE OBSERVATIONS FROM UARS MIS:
AN OVERVIEW OF ZONAL MEAN RESULTS

Lucien Froidevaux, Joe W. Waters, William G. Read,
Lee S. Elson, Dennis A. Flower, and Robert F. Jarnot

*Jet Propulsion Laboratory
California Institute of Technology
Pasadena, California*

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ABSTRACT

Global ozone observations from the Microwave Limb Sounder (MLS) aboard the Upper Atmosphere Research Satellite (UARS) are presented, in both vertically resolved and column abundance formats. We review the zonal mean ozone variations measured over the two and a half years since launch in September 1991. Well-known features such as the annual and semi-annual variations are ubiquitous. In the equatorial regions, longer-term changes tied to the quasi-biennial oscillation (QBO) are observed, with a strong semi-annual signal above 20 hPa. Equatorial ozone values near 50 hPa exhibit low values from October 1991 to June 1992, with a fairly rapid increase in early summer 1992, after which the low ozone pattern splits in two, yielding minima in the sub-tropics (possibly in connection with residual circulation changes tied to the QBO). The ozone hole development at high southern latitudes is apparent in MLS column data integrated down to 100 hPa, with a pattern generally consistent with Nimbus-7 TOMS measurements of total column; the MLS data reinforce current knowledge of this lower stratospheric phenomenon by providing a height-dependent view of the variations. The region from 30S to 30N (an area equal to half the global area) shows very little change in the ozone column from year to year and within each year.

The most striking ozone changes have occurred at northern mid-latitudes, with the October 1992 to July 1993 column values significantly lower than during the prior year. The zonal mean changes manifest themselves as a slower rate of increase during the 1992-93 winter, and some evidence for a lower fall minimum. A sub-tropical low starting in late 1992, with significant contribution from changes in the lowermost stratosphere (near 100 hPa) may play a role in the generation of low ozone at mid-latitudes. A recovery occurs during the summer of 1993; early 1994 values are significantly larger than during the 2 previous winters. These results are in general agreement with variations measured by the Nimbus-7 TOMS and Meteor-3 TOMS instruments at mid-latitudes. However, the southern mid-latitudes exhibit less of a column ozone decrease in the MLS data (down to 100 hPa) than in the TOMS column results, with possible implications on the altitude-dependence of the changes in both hemispheres. The timing and latitudinal extent of the northern mid-latitude decreases appear to rule out observed ClO enhancement in the arctic vortex, with related chemical processing and ozone dilution effects, as a unique cause. Local depletion from ClO-related chemical mechanisms alone is also not sufficient, based on MLS ClO data. The puzzling asymmetric nature of the changes probably requires a dynamical component as an explanation. A combination of effects (including chemical destruction via heterogeneous processes, QBO phasing, reduced ozone at low latitudes and subsequent poleward transport) probably needs to be invoked.

Finally, residual ozone values extracted from TOMS minus MLS column data are briefly presented as a preliminary view into the potential usefulness of such studies, with information on tropospheric ozone as an ultimate goal.

1. introduction

There have been various reports of stronger than expected declines in ozone during the last two years, from satellite measurements (Gleason et al., 1993; Herman and Larko 1994, Planet et al., 1994) to ground-based and balloon-borne data (Grant et al. 1992, 1994, Kerr et al., 1993; Bojkov et al., 1993; Hofmann et al., 1993, 1994; Komhyr et al., 1994). The potential for ozone destruction as a result of heterogeneous reactions occurring on volcanic (sulphate) aerosols has been increasingly discussed since the large eruption of the El Chichon volcano in 1982 and its possible relation to ozone changes (Hofmann and Solomon 1989). The possible impact of the June 1991 eruption of Mount Pinatubo on ozone is a subject of much current interest, given the large increase in stratospheric volcanic aerosol from that eruption (Bluth et al. 1992, McCormick and Veiga 1992, McCormick et al. 1994).

The Upper Atmosphere Research Satellite (UARS) was launched on September 12 1991, with 10 instruments of international origins aboard (Reber 1993). The study of upper atmospheric chemistry and dynamics, coupled with UARS measurements of solar flux and energetic particles, is at the center of this mission's research goals.

Ozone measurements from the Microwave Limb Sounder (MLS) are described in this paper, for the period between October 1991 and March 1994. We focus on the zonal mean variations observed throughout the globe during this interesting time. This overview is meant to present some of the more obvious features seen in MLS data, even though a full interpretation (with model comparisons) will have to await further studies. Since the MLS data

are now being released to the "non-USA" community, it is hoped that this paper can guide other interested scientists towards certain areas of research in relation to this data set. Elson et al. (1994) give an overview of the large-scale wave components of the MLS ozone data. Some words of caution and caveats are given in section 2, regarding retrieval uncertainties and known systematics, even though an overall good quality product has been achieved so far. The data set presented here was generated from MLS V3 Level 3 data files on the Central Data Handling Facility at the Goddard Space Flight Center; these are the files which are being stored on the Data Archive Accessing Center (DAAC), also at Goddard. Further details and profile intercomparisons with other data sets will be given in upcoming publications. In section 3, zonal mean ozone mixing ratios at various latitudes from 80S to 80N latitude are presented; some emphasis is placed on changes observed at low latitudes, and the possible connection with the QBO. The ozone abundances are then integrated in the vertical to produce zonal mean column measurements, for various pressure intervals; this is discussed primarily in section 4. We focus in section 5 on the ozone column behavior at mid-latitudes (30 to 60 degree bin) and the significant differences observed during the first two years of MLS operation, along with possible explanations for these measurements. (Comparisons with both the Nimbus-7 and Meteor-3 TOMS ozone column data are made. Section 6 gives a brief presentation of residual ozone column obtained from a subtraction of h41, S column from TOMS total column data, as a topic worthy of further study, regarding the possibility of extracting variations in tropospheric ozone.

2. MLS ozone retrievals

The MLS instrument measures thermal emission at millimeter wavelengths by scanning through the atmospheric limb (Waters, 1993). An instrument description has been given by Barath et al. (1993), and first results on polar ClO and ozone in the lower stratosphere are described in Waters et al. (1993a). Temperature (with tangent pressure registration from O_2 lines at 63 GHz) and water vapor (e.g. Harwood et al. 1993) are the other primary products, but information on SO_2 (Read et al., 1993) and HNO_3 (Santee et al., 1994) is also present in the spectra. Ozone is measured in two distinct spectral bands by the "205 GHz" and "183 GHz" radiometers and independent retrievals are carried out for each band. We report here on the 205 GHz results only, because they have shown somewhat better accuracy than the 183 GHz results and because they cover a longer time period (the 183 GHz radiometer measuring H_2O and O_3 stopped operating in mid-April 1993). Stratospheric abundances and variations are emphasized here (mesospheric information is mostly obtainable from the 183 GHz ozone retrievals). Atmospheric profiles retrieved in this fashion are spaced about 4 degrees apart in latitude, with better coverage near the latitudes corresponding to orbit turn-around. These turn-around points occur at about 34N (34S) and 80S (80N) when the satellite is flying forward (backward), with the alternating coverage arising as a result of the UARS yaw maneuvers (roughly every 36 days), tied to the orbit precession. There are 15 orbits per day, so that each latitude (at the 4 degree resolution) is sampled about 30 times during a 24 hour period.

This type of sampling goes into the zonal means discussed throughout this paper.

Substantial details of MLS ozone retrievals and comparisons with other data sets will be discussed in upcoming work, and continued validation and refinements in the retrievals are to be expected.

The MLS retrieval technique uses a sequential estimation approach (Rodgers, 1976) to obtain tangent pressure and temperature from the 63 GHz band, followed by mixing ratio retrievals in the other bands. Climatological *a priori* profiles, provided by UARS investigators (based on existing data sets, and models where data are lacking) are combined with the data in this technique, mostly for stability in altitude regions where measurement sensitivity is rapidly degrading; we have used large *a priori* errors to ensure that minimal bias is introduced in the retrievals. Given the linearity in the radiative transfer at these wavelengths, a single pass fit gives good results, and we have not used optically-thick channels. However, further improvements can be expected with a fully iterative retrieval, to be implemented for the next major reprocessing of the MLS data. The current retrieval mixing ratio profiles consist of joined linear segments having 3 equally spaced breakpoints per decade change in logP (i.e. at pressures of 100 hPa, 46 hPa, 22 hPa, ...), which corresponds to a vertical (Level 2) grid with roughly 6 km spacing. The atmospheric profiles are retrieved as the breakpoint values. An approximate interpretation of these values, relative to the true (infinite resolution) profile, is the least-squares fit of the linearly-segmented retrieval profile to the true one. The integrated area under the retrieved profile is conserved regardless

of the vertical structure in the real profile or the retrieval grid spacing. The Level 3 standard UARS grid is twice as fine as the MLS Level 2 grid, i.e. it has 6 points per decade in $\log P$ rather than 3. The MLS values at the non-retrieved Level 3 grid points are averages of the Level 2 coefficient values on either side. A trade-off exists between vertical resolution and profile noise; the optimum MLS vertical resolution is 5 points per decade in $\log P$, which is only slightly coarser than the Level 3 grid spacing. Profiles retrieved from MLS observations should generally be sensitive to features with vertical scale of a few km. Smearing effects from the antenna field of view and radiative transfer through the atmosphere lead to a vertical smearing of 3.2 km (4 km is often quoted as vertical resolution). Current estimates of retrieval precision ("1 sigma noise estimate") for single profiles are 0.3 ppmv between 1 and 4.6 hPa, 0.2 ppmv between 10 and 46 hPa, and 0.5 ppmv at 100 hPa. These figures are based on observed summertime variability (r.m.s. deviation about the mean) in latitude bands near turn-around (30-35N or 30-35S) - where a large number of profiles can be obtained in a narrow latitude range - as well as in the relatively quiet tropical regions (5S-5N); true precision may be slightly better than the minimum standard deviation obtained for the 8 days used in this analysis, but theoretical estimates agree well with this empirical method of estimating precision. For zonal means in a typical 5 degree-wide latitude bin, with about 40 to 45 individual measurements included, precision is expected to be of order 0.1 ppmv or less. In terms of the noise component on column ozone calculated by integrating the retrieved profiles in the vertical, as done in this paper, the correlation between levels

leads to some cancellation of errors. Our estimates of column precision, using the minimum observed variability in "quiet" periods/locations and near turn-around points (from a sample of 8 days, each with over 100 profiles in 2 separate latitude bins) are 7 DU (Dobson units), 2 DU and 1 DU for column ozone calculated above 100 hPa, 46 hPa, and 22 hPa respectively. For zonal means in a typical 5 degree-wide bin, these column precision estimates will go down by a factor of about six.

However, systematic errors will not "average out". Based on comparisons with other data sets, and based on examination of the fields themselves, we find that there are small biases at certain pressure levels. In particular, the 46 hPa MLS values may be somewhat low (a few tenths of a ppmv), whereas the 100 hPa values show an opposite bias (of order 0.1 to 0.2 ppmv high). The retrieval technique relies on fitting the spectral contrast within the instrument bandpass. In the lowermost stratosphere, this contrast is significantly reduced in comparison with the mid-to upper stratosphere, and it becomes difficult to separate from contributions (some better taken into account than others) from HNO_3 , H_2O , N_2O , dry air continuum, and other small instrumental or computational effects. We sometimes get zonal mean values which are negative at 100 hPa (the lowest level for reasonable MLS sensitivity), particularly in the summer months at tropical latitudes; this bias is not yet removed or completely understood. Finally, small oscillations in the mean field, tied to the UARS "yaw-cycle" - 36 day variation - , are known to exist (see figures later in the paper), particularly in the lowermost stratosphere; small discontinuities can also appear when a yaw day is crossed.

These features may be propagated to Ozone! through the retrieved tangent pressure effect, and further investigation is necessary to fully understand and remove these effects, which are present at the few percent level.

The above issues should not affect the main results presented here, which mostly deal with zonal mean trends. For example, excellent tracking has been obtained between MLS and SAGE II ozone mean values (D. Cunnold, private communication, 1993), despite a small offset between the two data sets (5-10 % higher values seen in MLS ozone), and other high-quality comparisons with ground-based, ozone sonde data and other UARS instruments have been made (to be published later). The MLS zonal mean radiances are generally fit by "forward model" radiances (using retrieved MLS fields) to within one percent (r.m.s.).

We now turn to a description of some of the interesting aspects of these 2.5 years of MLS data on zonal mean ozone.

3, Ozone mixing ratio data

Figure 1 is a time series representation of zonal mean ozone volume mixing ratio for 4 pressure levels (46, 22, 10, and 2 hPa), from October 1991 to March 1994. These zonal means are computed from MLS Level 3A 1₁ data (every 4 degrees in latitude). Each year of data (starting in October) is shown in a different color. Because of the UARS cyclic "yaw maneuver", MLS data alternate between mostly northern and mostly southern latitudes (coverage from 34S to 80N, followed by 34N to 80S). This leads to roughly 36-day gaps in MLS data at latitudes poleward of 34N or 34S (see Fig. 1). Other gaps

occasionally occur because of missing or bad data, caused by problems with the instrument or satellite. Figures 2 and 3 also display zonal mean MLS ozone mixing ratio data (for all retrieval points between 100 and 2.2 hPa, i.e. at 100, 46, 22, 10, 5, and 2 hPa), for each of the first 2 years of observation, as color contour plots. This brings out certain features more easily than in Fig. 1. Data used in these figures are derived from Fourier coefficients in time and longitude, as described by Elson et al. (1994), and accurately represent variations longer than one day.

There are a number of general points which one can make, based on the above figures, mostly as a check of previous results on stratospheric ozone. For example, largest ozone mixing ratios are observed at equatorial latitudes in the mid-stratosphere (see the 10 hPa plots), and largest variability is observed at high latitudes in winter, as expected. Note that small temporal oscillations can be seen (probably best in Figs 2 & 3 in the tropics), tied to the UARS yaw period of approximately 36 days. These are known artifacts in MLS data, and investigations continue regarding these effects, which are present at the few percent level. The lower stratospheric ozone maxima are observed during February/March in the northern hemisphere mid-to-high latitudes, with a similar peak during August/September in the southern hemisphere (see the 46 hPa plots); this spring maximum is caused by transport of high ozone air from the tropics, where the primary production occurs, with a subsequent decrease induced by increasing photochemical destruction (e.g. Perliski et al. 1989). The ozone hole-related decrease in ozone during August and September is observed at the highest southern lati-

tudes, in the 46 hPa plots (not so much in the 100 hPa plots, interestingly); a synoptic view of early results from MLS in the 1992 winter over these regions was given by Waters et al. (1993b). We will come back to the ozone hole briefly in section 4, but the polar regions are not the main emphasis of this paper; see Manney et al., 1994a,b, for recent discussions of polar ozone loss based on UARS data from MLS and the Cryogenic Limb Array Spectrometer (CLAES).

in the mid-stratosphere (see the 10 hPa plots) at mid-to high latitudes, the annual cycle dominates, as a consequence of significant photochemical production which maximizes in the summer. This annual variation is observed, with the expected 6-month shift between hemispheres. A well-known semi-annual oscillation (SAO) dominates at low latitudes in the mid-to upper stratosphere, as observed in these data as well (see also Eluszkiewicz et al., 1994). The SAO and associated vertical motions are believed to play a role in producing features like the double-peak structures in pressure/latitude cross-sections of SAMS N_2O and C_{11}H_4 fields (Gray and Pyle, 1987; Choi and Holton, 1991). In the upper stratosphere, temperature-dependent ozone destruction cycles play a dominant role, and the 2 hPa MLS plots appear to follow such trends (e.g. spring mid-latitude decreases when temperatures are increasing). These and other features related to the annual and semi-annual ozone variations are in general agreement with previous analyses by Perliski and London (1989) and Perliski et al. (1989). Further correlative studies would be useful, however, for quantitative conclusions on these variations. Ray et al. (1994) provide a more detailed analysis of the SAO, based on MLS data,

and comment on the amplitude characteristics versus pressure/latitude.

An interesting tropical feature is the existence of low ozone abundances at 46 hPa from Oct. 1991 to mid-1992. The low equatorial values (Fig. 1) rise sharply during June-July 1992 and level off until the Sep. 1993. From late 1993 on, low values are again observed near the equator. Fig. 2 further shows that this low ozone tropical feature splits into two somewhat weaker sub-tropical lows after the summer of 1992. The low ozone values may be linked to upward motion in connection with the quasi-biennial oscillation (QBO). Indeed, the sub-tropical lows occur during a period of equatorial westerlies (near 20 hPa), a period traditionally associated with enhanced downward motion at the equator, with upwelling in the sub-tropics as a result of the return arms of the induced circulation (e.g. Gray and Pyle, 1989). Observations of the sub-tropical ozone QBO, as deduced from TOMS data by Bowman (1989), can place constraints on details of the mechanism for the QBO and the spread of related anomalies to other latitudes. We simply note here that the sub-tropical low ozone values are reasonably symmetric about the equator - Bowman (1989) and Lait et al. (1989) point out that the TOMS analyses show a more symmetric QBO behavior about the equator than previous analyses of ground-based or Nimbus-4 BUUV data -. However, uplift effects and circulation changes arising from post-Pinatubo aerosol heating would also have to be considered as an explanation for low tropical ozone (Kinne et al. 1992, Schoeberl et al. 1993, Grant et al. 1992, 1994, Pitari 1993). Subsequent spreading of the aerosol to higher latitudes would also need to be taken into account; SAGE II aerosol data for example, have pro-

vided much information about the poleward dispersal of Mount Pinatubo aerosol, a process itself partly connected to the QBO (Trepte and Litchman 1992; Trepte et al. 1993). The time constants for lofting (Kinne et al. 1992), and the fairly symmetric nature of the latitudinal cross-sections of aerosol extinction measured by ISAMS (Lambert et al. 1993) and CLAES (Mergenthaler et al. 1993) during 1992 would lead us to believe, however, that the MLS-observed splitting of tropical low ozone values into the sub-tropics is a result of residual circulation effects, possibly tied to the QBO, as opposed to aerosol-induced lofting effects. We examine further in Fig. 4 the variations in equatorial mixing ratio (ΔO_3) with respect to the mean over the two and a half year period of MLS observations. This figure shows that the changes mentioned above for 46 hPa are somewhat anti-correlated with changes occurring at 100 hPa. We keep in mind the caveats mentioned in section 2 on error estimates, with a remote possibility that unmodeled variations in HNO_3 or upper tropospheric H_2O could play a role in the ozone effects seen here; however, we feel that the main characteristics shown here are actual changes in atmospheric ozone. The suggestion from the anti-correlated behavior in these lower stratospheric levels is one of oppositely directed vertical motions at the 100 and 46 hPa levels. We do not know whether the MLS variations for 100 hPa shown in Fig. 4 are consistent with model expectations at these levels, but the expected opposite vertical motions on either side of the zero-wind line (in relation to the QBO westerly/easterly regimes) could play a role in such variations. We also note that the 100 hPa values appear to be more affected by annual variations than the 46 hPa values. The numeri-

cal simulation of the ozone QBO by Gray and Dunkerton (1990) exhibits a 6 month phase shift between the maximum in ozone at altitudes below 23 km and the maximum at about 26 km, which the authors attribute in part to chemical control (NO_y variations) at the higher altitudes. The observed phase shift between the observed steep rise in MLS ozone at 22 hPa and at 46 hPa appears to agree with that type of behavior. With more MLS data and further analyses, altitude and latitude dependent results on the ozone QBO (amplitude and phase) could be obtained, as done for SAGE II data by Zawodny and McCormick (1991). In the mid-to upper stratosphere, observed variations displayed in Fig. 4 show a transition towards changes dominated by a semi-annual signal (at 10, 5 and 2 hPa), with noticeable phase changes at the different levels.

Pursuing the equatorial variations a little further, we calculate the ozone column by integrating the mixing ratios in the vertical, using the retrieved MLS profile points only (i.e. every other Level 3 grid point). Then, based on the MLS Level 3A 1 files, we produce an area-weighted average for the 5S to 5N latitude bin. In order to study the height-dependence in the column amounts, the column ozone values above 100, 46, and 22 hPa are plotted in Fig. 5 as a function of time since October 1, 1991. A 36-day smoothing (running average) has been applied to help remove the known spurious oscillation tied to the UARS "yaw period". While the column above 22 hPa displays a strong semi-annual oscillatory behavior, clearly connected to the mixing ratio variations of Fig. 4, the column down to the lower levels (100 and 46 hPa) is also affected by a longer-period variation. We infer that this

must be a manifestation of the quasi-biennial oscillation in column ozone [see e.g. Oltmans and London, 1982]; a definite characterization of the QBO from MLS data would require a longer time series and removal of the mean annual and semi-annual components. More thorough analyses from satellite data have been performed (e.g. Hilsenrath and Schlesinger 1981, Hasebe 1983, Bowman 1989). Nevertheless, it is hoped that further investigation into the 46 and 100 hPa ozone data from MLS (see fig. 4) will lead to an improved understanding of the QBO signal and its generation. The bottom panel of Fig. 5 shows the zonal mean winds at the equator, at 46 hPa and 22 hPa, based on United Kingdom Meteorological Office (UKMO) data (Swinbank & O'Neill 1994). The ozone QBO has traditionally been linked to the lower stratospheric wind QBO and wave-driven vertical motions (Holton and Lindzen 1972, Plumb 1984) which can modulate the total column. Lait et al. (1989) have found better correlation between TOMS total column ozone QBO signal and the 30 hPa zonal winds at the equator, than with the 50 hPa winds. Also, Bowman (1989) finds maximum correlation between TOMS column ozone data and zonal mean winds at 20 hPa. This agrees with the general relationship shown in Fig. 5, where better correlation is apparent for winds at pressures of 22 hPa than at 46 hPa. The maximum ozone values occur during the westerly phase of the dynamical QBO near 22 hPa, presumably in connection with the downward motion generally ascribed to this QBO phase (e.g. Hamilton, 1989; Gray and Dunkerton, 1990). We note that the long-term column ozone variations seen in Fig. 5 (for the columns down to 100 and 46 hPa) are mostly driven by the changes at 46 hPa shown in Fig.

4, in qualitative agreement with the implications from Gray and Dunkerton (1990). Furthermore, the exact phase relationship between ozone and zonal mean winds at various heights can be affected by the feedback mechanism of ozone heating on the circulation, as well as by photochemical effects [see the recent work by Masche (1994) and earlier modeling by Ling and London (1986)]. Further comparisons with such models would be of much interest, despite the relatively short time span of MLS observations.

We now extend the consideration of column ozone variations to other latitudes, given the importance of the column to attenuation of solar UV radiation and the existence of long-term measurements of column ozone from other instruments.

4. An overview of MLS ozone column data

An analog to the mixing ratio time series of Fig. 1 is shown in Fig. 6, for ozone integrated above 100 hPa, every 16 degrees in latitude. The interannual ozone column changes, for this two and a half year time period are seen to maximize in the northern mid-] latitudes. In particular, the 48N October values are very similar for the three years, but the rate of increase towards the well-known March maximum varies substantially in each of these years. The second year (solid dark blue line) shows lower values than the first year (dotted light blue line) in the winter, while the third year (thick red line) exhibits ozone values higher than the first year in that season. It turns out that most of these changes are driven by interannual variations in the 100 hPa mixing ratios, as discussed further below. The July through

September column ozone values are quite similar during 1992 and 1993 for mid-to-high northern latitudes. The equatorial column behavior mirrors the changes seen in Fig. 1 at 46 hPa, as shown also in the previous section. Interannual variations at the southernmost latitudes (80 degrees south) are most apparent during the vortex break-up phase. (November), as one might expect and are quite small at other times.

In order to get a broader view of the changes at different latitudes, we have produced area-weighted integrals of the column ozone variations for different latitude bands in Fig. 7 (top left panel), as opposed to the specific latitudes of Fig. 6. The globe has been split into latitude intervals ranging from (in degrees) 0 to 15, 15 to 30, 30 to 60 (mid-latitudes), and 60 to 80 (polar latitudes); also the 30 S-80N, 30S-30N, and 30N-80S bins are shown. The vertical intervals are the same (100 DU from minimum to maximum) except for the 60-80S bin. Furthermore, Fig. 7 includes three other panels to give column above 22 hPa's, and the column amounts between 100 and 46 hPa, and between 46 and 22 hPa. Again, the striking feature is the northern mid-latitude (30-60N) wintertime column decrease (or slower rate of increase) during 1992-93 versus 1991 -92; an average drop of about 8% (20 to 25 DU) is evident from mid-December to mid-March (if one assumes continuity in the time interval during which MLS is not viewing these regions). Roughly half of this decrease arises from the layer between 100 and 46 hPa, and another quarter comes from the 46 to 22 hPa layer, as can be seen from an examination of the other panels in Fig. 7. Similarly, more than half of the recovery in the 1993-94 winter is coming from the lowermost layer measured

here (roughly 50 to 100 mPa). The 1993 decrease is reduced in May, and has essentially disappeared by mid-summer. In early 1994, column ozone at northern mid-latitudes is roughly 5% higher than in early 1992. The highest northern latitudes also show larger ozone values in late 1993, but values close to the early 1992 measurements in early 1994. While the 1992-93 changes in the 60-80N latitude bin are similar to those occurring in the 30-60N bin, they are, interestingly, smaller in magnitude; overall, this is also true of the changes in the 15-30N bin. We discuss the mid-latitude ozone observations and potential implications at greater length in section 5.

In the tropics, the second year of MLS observations generally shows larger ozone column values than the first year, in contrast to the behavior at higher latitudes in the north. The integral over 30S to 30N (an area equal to half the global area) shows very little change from year to year and within each year in the ozone column above 100 hPa.

The other fairly large difference of note is the October-November 1992 low ozone in the 15-30S latitude bin; in this case, an examination of the mixing ratios shows that these changes appear to come mostly from the pressure levels above 100 hPa (46 hPa in particular). A large component (about half or more) of the 15-30 degree bin column ozone values and annual variations (in both hemispheres) comes from the column above 22 hPa. The average ozone column changes in the 30-60S bin from year to year are significantly smaller than the average decrease in the corresponding northern hemisphere bin. Finally, the 60-80S latitude bin clearly shows the existence of an ozone hole development during August/September and the largest interannual vari-

ability is observed during vortex break-up in November.

More focussed descriptions of the ozone changes in the polar vortices, as observed from MLS data analyses have been given by Waters et al. (1993a,b), and Manney et al. (1993, 1994a,b). In particular, Manney et al. (1994a) discuss the evidence for vortex-averaged ozone depletion in the northern winter, by contrasting the behavior of ozone with long-lived tracers measured by CLAES. Although we see no evidence for an ozone hole development in the zonal mean ozone column during the winter at 60–80N, as observed in the 60–80S bin during September, the rate of increase in the column is no doubt reduced by chlorine-induced ozone destruction (given the MLS ClO observations also presented in some of the above references).

We conclude this section with a comparison of the MLS ozone variations at high southern latitudes with Nimbus-7 TOMS total column data. Fig. 8 gives a height-dependent view of the column changes as seen by MLS, for column above 100, 46, and 22 hPa, during the months of June through November 1992. This shows that the overall trends observed by TOMS are fairly well reproduced in the MLS column data above 100 hPa, both during the decline and the springtime increase. Exact tracking between these data sets is not expected, because of the lack of polar night data in the case of TOMS and because of the different horizontal and vertical resolution between these instruments (TOMS can sample essentially down to the ground, if the effects of clouds and tropospheric profile assumptions on the retrievals are small). It is not quite yet clear, though, why better correlation does not exist in the short-term peaks occurring in September (these are tied to warming

events; see Fishbein et al. 1993). The figure also adds MLS information on column above other levels, and this shows that most of the depletion in total column comes from a reduction in the ozone amount between 100 and 50 hPa, in agreement with previous finer resolution vertical profiles from ozone sondes over Antarctica (e.g. Hofmann et al. 1989). In contrast, the column above 22 hPa is observed to increase during this time period, presumably because of a combination of production increase (more sunlight) and mixing of higher Ozone! values from lower latitudes.

5. Mid-latitude ozone decreases

a. *Further observational analyses*

We pursue here a more detailed description of the mid-latitude ozone decreases in 1992-93, as well as possible implications. First, a comparison with the TOMS results is warranted. Nimbus-7 TOMS stopped functioning in May 1993 (last good full day of data is May 6), after over 14 years of outstanding operation. We have added recent results from the Meteor-3 TOMS instrument as comparison for 1993, including the 1 January to 6 May overlap period with Nimbus-7 TOMS. The Meteor-3 instrument is in a precessing orbit which leads to periods of time during which observations over a good portion of a hemisphere are not possible (solar zenith angles are too high); because of this (and instrument malfunction during most of June 1993), we have omitted all of June at all latitudes, as well as all of July for southern latitudes from the Meteor-3 data record. The TOMS and MLS column amounts from October 1 1991 to end of September 1993 are shown in Fig.

9, for the 30-60 northern and southern latitude regions. We see from the figure that very good agreement is obtained between both TOMS instruments during the 1993 overlap period shown here. The 1992-93 decrease in ozone column is evident in TOMS data (as discussed by Gleason et al. 1993). This is particularly true during winter/spring; in the north, decreases in the south are largest in the early part of the diagram, i.e. in October/November. By end of August 1993, TOMS average ozone column amounts have basically recovered to the 1992 values, but may still lie somewhat below the expected lows based on long-term variability (Herman et al. 1994); these authors note that the average symmetry between hemispheric (long-term) decreases has been broken since the onset of large ozone losses in 1992, mostly in the northern hemisphere. The MLS ozone column data appear to agree qualitatively with the general results seen by the TOMS instruments over this time period. Large northern hemisphere decreases are also seen by MLS, especially in the wintertime, when the absolute decrease is similar in both data sets (20 to 25 DU). Recovery to 1992 values appears to occur somewhat quicker in the MLS results than in the TOMS data. The recovery in the south (in August) is observed in both data sets. The main difference is in the magnitude of the southern decreases; TOMS-inferred decreases are significantly (50 to 100 %) larger than the MLS column differences during November and January/February in particular. Keeping in mind the MLS column precision estimates in section 2, and the fact that a large number of profiles (more than 250) are averaged in the latitude bands discussed here, noise in the MLS averages should be at the sub-Dobson unit level. Systematic effects (see also

section 2) can remain, as seen for example in the artificial oscillations for MLS data within each UARS yaw period. These effects (of order 5 DU) are reproduced in both years, however, and should not affect the interannual differences at the systematic levels (more than 10 DU) observed by TOMS in the south. Rather, a systematic interhemispheric difference between both years would have to be invoked, in either TOMS or MLS data. Taken at face value, the differences could imply that the column ozone decreases in the southern hemisphere occurred at relatively lower altitudes (with respect to the 100 hPa pressure level) than in the northern hemisphere, so that MLS did not sample the whole effect in the south. It appears unlikely that aerosol or tropopause height effects could cause such differences in the retrievals between TOMS and MLS measurements, and further investigation into this discrepancy in the magnitude of the interhemispheric differences is needed. Unfortunately, there are fewer ground-based or ozone sonde data sets in the south than in the north to help in this matter.

We offer a finer resolution picture of the MLS column changes in Figure 10, where the complete zonal mean column data set is mapped as a function of time, for the two years discussed above. The bottom panel gives the difference (second year minus the first) and puts the temporal and latitudinal extent of the changes at northern mid-latitudes during the first part of the time period (wintertime in particular) in evidence. One of the changes contributing to the observed mid-latitude year-to-year differences can be found by examining the 20-30N latitude region, where the depth of the annual minimum in the fall is seen to be larger in the second year (1992). Also, the 30-40N band exhibits a

decrease between October and December, as opposed to the increase in the previous year. The low ozone values in the 20-40N latitude region arise as a combination of the low values at 46hPa, observed in the splitting of the equatorial low mentioned in section 2, and the overall lower ozone values in the 100 hPa region (see Fig. 2).

Panels in Fig. 11 depict the changes in latitude/pressure cross-sections from early December through mid-March, in roughly two week averages. The first year (1991-92), second year (1992-93), and differences between the two are shown. Ozone concentration (units of 10^{12} molecules/cm³), rather than mixing ratio is plotted, to emphasize the lower stratosphere, where changes at 46 and 100 hPa are most important for column abundances. Higher equatorial ozone values are observed near 46 hPa in the second year, as seen clearly in the difference plots, and the early December southern sub-tropical decrease is also seen. The effect of lower ozone concentrations in the 100 hPa pressure range in particular is evident at mid-latitudes, with the differences migrating to higher latitudes as time progresses. In late February and early March, the ozone decreases are spread over a larger vertical extent than during December, possibly as a result of polar vortex processing and subsequent dilution effects at lower latitudes.

b. Discussion

Figure 11 is consistent with the long-held views that high latitude winter-spring ozone column values arise from the poleward and downward transport of large ozone concentrations produced in the tropics. The mid-latitude fall-winter decrease may be related to the mixing of lower than usual values from

the 50-100 hPa vertical range at low latitudes (20-40N) into the lowermost (100 hPa) mid-latitude regions. Polar region decreases may have played a role in February/March [indeed, higher ClO is observed by MLS in the vortex in 1993 (Manney et al. 1994a)], but not in the early winter, when high latitude changes between the two years are small and much lower ClO is present in the vortex. Indeed, MLS results from the previous section show that the ozone decrease in 1992/93 does not start in the polar regions and spread to lower latitudes.

The low ozone values observed at mid-latitudes by MLS are generally consistent with other recently reported measurements such as the Nimbus-7 TOMS results (Gleason et al. 1993), indicating unprecedented low global ozone in 1992, the TOMS update by Herman and Larko (1994) with HETE-3 TOMS data added up to July 1993, and other ground-based and satellite results (Bojkov 1993, Kerr 1993, Hofmann et al. 1993, 1994, Komhyr et al. 1994, Planet et al. 1994). Given the TOMS historical record and statistical models which can fit this record up to 1991, the 1992 and 1993 ozone column changes at northern mid-latitudes in particular are significantly outside expectations (Gleason et al. 1993, Herman and Larko 1994). Despite the difference in the magnitude of the hemispheric asymmetry in the decreases discussed above for MLS versus TOMS, the MLS data support the statements by Herman and Larko (1994) that "there is no longer an average symmetry between the hemispheres", based on the 1992/93 data and comparisons to long-term records (Stolarski et al. 1991, 1992, Herman et al. 1993, McCormick et al. 1992, Niu et al. 1992). The 1994 MLS data appear

to show a return to pre-1992 values, which should be more in-line with the historical envelope discussed in TOMS data analyses such as those of Herman and Larko (1994). The northern mid-latitude ozone decreases observed during the two years after the eruption of Mount Pinatubo (June 1991) are about an order of magnitude larger than the already significant long-term decreases discussed in some of the above references. There were also equatorial ozone decreases (Grant et al. 1992, 1994, Schoeberl et al. 1993, Herman and Larko, 1994) in the months just after the eruption. The low MLS equatorial values (late 1991 and early 1992) appear to support the latter findings, but the MLS data lack the long-term record for comparisons from the pre- and post- Pinatubo time periods. Indeed, other effects probably played a role in the continuation of low ozone at low latitude, as observed in MLS data.

The combination of these observed ozone decreases, as well as the recovery in late 1993, point to Mount Pinatubo as a likely culprit for at least part of the ozone changes. As mentioned above, as well as by Herman and Larko (1994), the timing of the decreases does not support a polar processing/dilution effect alone. Furthermore, known natural causes of ozone decrease such as solar flux variations or QBO effects appear to be too small to explain, by themselves, the large recent changes in ozone (Schoeberl et al. 1993, Chandra 1993, Gleason et al., 1993, Herman et al. 1993). Rather, a combination of effects is probably required, but the timing, duration, magnitude, and latitudinal extent of the ozone decrease all need to be addressed. Heating by the volcanic aerosol appears to have occurred after the Mount Pinatubo eruption (Labitzke and McCormick, 1992). Such heating can then lead to uplift

(and subsequent adiabatic cooling) in the tropics] lower stratosphere (Kinnison et al., 1992), with accompanied reductions in the ozone concentration. Further changes in the circulation can also be expected, and some models have addressed these issues (Brasseur and Granier 1992, Granier and Brasseur, 1992, Pitari 1993, Pitari and Rizi 1993, Kinnison et al. 1994), even though a fully coupled three dimensional simulation of the radiative, dynamical, and chemical effects (with realistic aerosol distribution and decay) is a difficult task. Radiative effects alone, however, would lead to a decrease in the tropical ozone amounts, but an increase in the mid-high latitudes (based on such models). Thus, increased chemical destruction associated with heterogeneous reactions on the sulphate aerosols appears to be needed to explain the mid-latitude ozone losses. Such effects were discussed previously (Jöreskog and Solomon, 1989) in connection with the El Chichón eruption and possibly related ozone decreases. In particular, N_2O_5 hydrolysis on sulphate aerosols, and the related decrease in available NO_x and increase in ClO_x (and HO₂, has been implicated as a major reason for long-term trends in ozone (Rodriguez et al. 1991), as well as for observed changes in lower stratospheric species concentrations (e.g. Johnston et al. 1992, Halley et al. 1993, Avallone et al. 1993, Solomon et al. 1993, Kawa et al. 1993); aerosol-induced changes in photolysis rates can also cause perturbations in chemical species abundances (e.g. Michelangeli et al. 1989, 1992). However, the early reduction in the tropics is likely a result of changes in the atmospheric heating and circulation (e.g. Brasseur and Granier, 1992).

Model calculations of the impact of heterogeneous reactions on sulphate

aerosol (see also Pitari et al. 1991) generally predict largest ozone decreases at high latitudes. A recent 2-D model with more realistic aerosol distribution by Rodriguez et al. (1994) gives a reduction (from a background aerosol case) of about 2-4 % in the 30-60 degree latitude region, with a peak at latitudes poleward of 60 degrees. Reasons for the high-latitude peak include the larger availability of inorganic chlorine, the longer replenishment time for ozone, and the slower photodissociation rate of JNO_3 produced from the heterogeneous reactions (see also Hofmann and Solomon 1989). Interestingly, ozone destruction cycles involving enhanced HO_x amounts play a larger role than cycles involving the enhanced ClO_x abundances.

The following aspects of the MLS (and TOMS) observations may be the most challenging to understand in detail: (1) the latitudinal extent of the changes, i.e. the observations do not support a high-latitude maximum decrease generally predicted by models (2) the asymmetry in the decreases about the equator, and (3) the large decrease in 1993, occurring later than the year directly following the eruption. Issue (1) may have to do with the model treatment of poleward (and cross-vortex) transport of the aerosols and related heterogeneous chemistry effects. The extent of recent ozone changes at northern latitudes resembles more the results of Pitari et al. (1991) if the aerosol distribution is artificially limited to latitudes south of 55N (their Fig. 2b), but this may be an unrelated coincidence. If enhanced chemical destruction is taking place at high latitudes as a result of volcanic aerosol, enhanced poleward transport of ozone from higher tropical altitudes could mask the effect. It is interesting, that the long-term ozone trends based on

TOMS data, as deduced by Niu et al. (1992), show mostly negative values at high latitudes, but some longitudinal regions of increase as well, during the winter only. For issue (2) above, it is worth noting that enhanced northward transport is not unexpected (see Pitari and Rizvi 1993), after the eruption of Mount Pinatubo. How this north-south asymmetry could lead to a larger decrease in ozone in the northern mid-latitudes (as opposed to the high latitude effect mentioned above) is another issue. Details of the transport in the lowermost stratosphere (and possibly the troposphere) could play a role here. For example, Hou et al. (1991) have shown that effects ranging from planetary waves (see also Leovy et al. 1985), to mesospheric drag and tropospheric forcing (in addition to photochemistry) can influence the lower stratospheric ozone amounts (and the column ozone); they also discuss the north-south asymmetry in transport (in relation to the asymmetry in the spring column maximum). The aerosol observations from SAGE (McCormick and Veiga 1992, Trepte et al. 1993) have shown that the Mount Pinatubo aerosols spread mostly southward in the first months after the eruption; however, the altitude distribution of the northward and southward components was different, with the northward dispersion occurring at lower altitudes. These lowermost stratospheric altitudes may be the critical difference, based on MLS ozone data in both hemispheres. However, the UARS aerosol extinction data from ISAMS and CLAES in 1992 do not show much asymmetry between the two hemispheres (Lambert et al. 1993, Mergenthaler et al. 1993). In terms of the chemical effects, Deshler et al. (1993) note that at 41°N latitude, the important aerosol surface area (and mass) showed a maximum about 6

months after the eruption, with a fairly steady behavior through 1992. We add that the mid-latitude ClO data from MLS show little asymmetry between the hemispheres (Waters et al. in preparation). However, the data in the lowermost stratosphere have the largest errors and could mask the existence of some north-south asymmetry there. Also, in connection with question (3), the MLS ClO data make it difficult to explain the 8% reduction in ozone during the winter of 1992-93 by [YO-related chemistry alone (if we use first-order estimates of the impact of the ClO-dimer, ClO-BrO, and ClO-HO₂ rates on ozone column); this is also true for more complete calculations (including HO_x and other cycles) by Rodriguez et al. (1994), who find a few percent reduction in the column. The latter authors point out that saturation of the N₂O₅ hydrolysis heterogeneous reaction occurs during both years following the eruption, and this - combined with the long time constant for ozone in the lower stratosphere - explains the long steady duration of the chemical destruction. Given the magnitude of the observed changes, we find it likely that other effects played a role as well]. Although Rodriguez et al. (1994) note that uncertainties in heterogeneous reaction rates could be invoked, this would not resolve the question of an asymmetry between hemispheres (based on their model). Hofmann et al. (1994) point to the QBO as only a partial possible explanation for the larger decreases in ozone observed in ozone sonde data during 1993. Indeed, the phasing of the QBO (see Chandra 1993, Zerefos et al. 1992) would imply a relative high in mid-latitude ozone in early 1992, followed by a relative low in early 1993 (almost opposite in phasing to the variations shown here at the equator for h41,S

column ozone). The strength of planetary-scale waves is also coupled to the phase of the QBO (Holton and Tan, 1980). Based on the MLS data shown in this paper, the northern hemisphere sub-tropical low ozone column during late 1992, and subsequent mixing; to higher latitudes during winter - in the lowermost stratosphere - , could play a part in the low northern mid-latitude ozone values during the 1992-93 winter. If the mid-latitude lower stratospheric ozone values during winter occur largely as a result of transport from higher altitudes in the tropics, however, less efficient transport in 1992-93 would have to be invoked. Previous models (e.g. Tung and Yang 1988) have addressed seasonal changes in column ozone, particularly at high latitudes, but have not emphasized the seasonal behavior of mid-latitude ozone in the lowermost stratosphere.

5. Residual ozone (TOMS minus MLS)

Since the ozone data from TOMS give the total column down to the ground (under the caveats mentioned earlier) and are generally quite consistent with ground-based measurements of total column, one is tempted to subtract stratospheric column ozone values from the TOMS data in order to infer tropospheric ozone. Ozone in the troposphere is a precursor for OH, an important radical which controls the oxidization of various source gases, including methane and hydrogenated chlorofluorocarbons (HCFCs). Using TOMS and SAGE data, Fishman et al. (1990) found that residual (TOMS minus SAGE) ozone was large downwind of Africa in the tropics and that the abundances maximized during the time of biomass burning (the dry sea-

son), between July and October. similar results were obtained by Croset al. (1992), from an analysis of the satellite data over Brazzaville, Congo (4S latitude); tropospheric ozonesonde data from that site appear to agree fairly well with the satellite residual values. Tropospheric ozone column amounts in the latter study range from about 30 to 50 DU, with a peak in late summer. Furthermore, the observed seasonal cycle in tropical total ozone column may be dominated by tropospheric ozone variations, (Oltmans 1981, Logan and Kirchhoff 1986, Fishman et al. 1986); the amplitude of the seasonal cycle in the tropics is typically of order 20 DU and the ozone maximum at the surface tends to occur about two months earlier than the stratospheric ozone peak, based on limited data sets (Oltmans 1981). There is considerable interest in determining the controlling factors for tropospheric ozone (biomass burning, for example, versus stratospheric intrusions); the model by Law and Pyle (1993) lends some support to the existence of phase differences between the troposphere and the lower stratosphere in the tropics.

We show here an example of residual ozone from Nimbus-7 TOMS minus MLS column ozone (where the MLS column is above 100 hPa). The latitude bin chosen here is 5 to 10 degrees south, in a region studied by some of the above authors, because of the existence of heavy biomass burning during July-October. We have looked at several other latitude bins, notably between 30S and 30N since the gaps in MLS data at higher latitudes make the analyses somewhat weaker; the 5-10S latitude bin gives one of the cleanest residuals (keeping in mind the artificial yaw-period oscillation in h41,S data, which can be detected in this figure). The residual ozone has a clear seasonal

signature. The amplitude of the variation and the timing of the residual maximum coincide reasonably well with results shown in the above references. It is also interesting that a phase shift is observed between the residual calculated in Fig. 12 and the residual calculated using MLS data down to 46 hPa only (see Fig. 13). This could be interpreted as a phase difference occurring as a result of the different seasonal peaks in the troposphere and stratosphere (since the curve for 46 hPa is more influenced by stratospheric ozone). Preliminary analyses at other latitudes do not yield such a distinct phase change. While these preliminary results may have some significance in terms of tropospheric ozone, we prefer to leave them - in this overview paper - as an indication of potentially interesting signals, which must be interpreted with great care. The existence of HIRS systematics at the 5 DU level, along with the interpretation of TOMS data (e.g. in the presence of clouds), indicate the need for further detailed analyses. An updated version of MLS data, with improvements in the lowermost stratosphere, is *planned*. Despite caution about the current data and the desire not to make premature statements about "tropospheric Ozone" here, we suggest that the MLS data set, coupled with TOMS ozone column, has the potential for inferring valuable information on this interesting topic.

7. Conclusions

We have reviewed the observed variations in MLS stratospheric ozone data, based on analyses of two and a half years (October 1991 to mid-March 1994) of zonal mean mixing ratios and column abundances. Expected periodic variations are clearly observed; for example, we note the existence of

a clear SAO in tropical ozone, for pressures less than about 20 hPa, and the dominant annual cycle at mid-high latitudes. Interesting variations are measured in the tropical lower stratosphere: a low equatorial ozone feature near 50 hPa, which splits into two sub-tropical lows during the summer of 1992, and reforms a single branch during the fall of 1993. A somewhat anti-correlated variation is observed at 100 hPa, but the existence of a systematic negative bias in the summer tropical values needs to be kept in mind. Given the time constants for the maintenance of aerosol lofting effects (Kinne et al. 1992), and the observed dispersal of aerosols, we would tend to ascribe these lower stratospheric variations to changes in the residual circulation, possibly Q130-related (vertical motions leading to lower ozone), as opposed to an aerosol-induced change. MLS ozone column data comparisons with TOMS data in the southern high latitudes (60°S-80°S) exhibit seasonal trends (for June through November) consistent with the overall TOMS variations; the ozone hole development is shown to occur primarily as a result of decreases in the 100 to 50 hPa region (in agreement with other data); based on MLS zonal mean data, the decrease is primarily driven by changes near 50 hPa. The largest ozone column changes from year to year have occurred during the December to March time frame at northern mid-latitudes, with the 1992-93 wintertime values typically 8% lower than the corresponding 1991-92 abundances. This slowing in the rate of ozone increase during the winter months appears to be related in large part to decreases in the lowermost stratosphere (near 100 hPa) during early winter, with an associated poleward spread, rather than a polar effect which migrates south; however, dilution ef-

fects from chlorine-related processing of polar ozone may have contributed to the decreases during February-March 1993, based on the significantly enhanced ClO values observed in the arctic vortex at that time (Manney et al. 1994a). While the phasing of the QBO is expected to lead to low ozone at mid-latitudes in late 1992, compared to early 1992, this effect alone would not be expected to be large enough to explain the observations. Since models do not predict that radiative/dynamical changes could - by themselves - lead to the type of mid-latitude behavior observed, it is likely that chemical cycles causing ozone destruction as a result of heterogeneous chemistry on volcanic aerosols have played a role in the observed decreases, as predicted by various models. However, the latitudinal extent of the changes (maximum decrease is not at high latitudes) and the asymmetry in the decrease (northern mid-latitudes significantly more affected than the southern mid-latitudes) are not easy to understand, given the known characteristics of the aerosol dispersion after the eruption (and knowledge of the mid-latitude ClO data from MLS, Waters et al. in preparation). We note that the TOMS-inferred asymmetry is not as large as the MLS-deduced effect; this could mean that changes in the southern latitudes occurred somewhat deeper in the atmosphere, but this also requires further investigation. One mechanism which could, in principle, explain the asymmetry, would be poleward mixing of low tropical ozone values, with stronger mixing towards the northern mid-latitudes; however, we do not know if the details of such a mechanism are plausible. What is needed, it seems, is an understanding of the changes in the lowermost stratosphere. A detailed analysis of other chemical species (or dynamical tracers)

would be desirable, possibly from the Cryogenic Limb Etalon Spectrometer (CLAES) data set (even though these observations ceased in early summer 1993). An examination of HNO_3 or NO_x fields (in addition to the ClO from M 1S) may also shed some light on the magnitude of possible changes caused by heterogeneous chemistry.

The low ozone values observed by M1S and other instruments from late 1991 to mid-1993 appear to have essentially disappeared during the late fall 1993 and early winter 1994. This recovery also seems to imply that effects related to the eruption of Mount Pinatubo were involved in the temporary ozone depletion, in combination with QBO and other effects. We expect that future analyses of the M1S data, in conjunction with others from UARS and elsewhere, will continue to educate us about the particularly interesting atmospheric experiment which seems to have occurred during the UARS mission.

Finally, analyses of the TOMS minus M1S residual ozone column should eventually improve our understanding of tropospheric ozone variations, and this paper's brief presentation of residual ozone gives a preliminary indication of the potential use of these combined data sets.

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FIGURE CAPTIONS

F] G. 1. Zonal mean ozone volume mixing ratio time series for October 1 1991 through March 14 1994, as measured by MLS. Retrievals for four different retrieval pressures are chosen: 46 hPa (bottom left), 22 hPa (bottom right), 10 hPa (top left) and 2 hPa (top right). These data are from MLS Level 3A files, gridded every 4 degrees in latitude. Time series are shown from the equator to 80 degrees latitude (the maximum latitude observable by MLS), in 16 degree increments. Oct. 1991 through Sep. 1992 values are given by light blue dotted line, the following year is given by dark blue solid line, and the Oct. 1993 through mid-March 1994 period is shown by red solid line. Gaps in the time series occur at mid-high latitudes because of the alternating coverage between north and south (see text).

FIG. 2. Stratospheric zonal mean ozone mixing ratios from MLS data created using the synoptic mapping technique (see Elson et al. 1994), starting with MLS Level 3A (time-ordered) files. Contour plots are shown as a function of latitude and time, for first two years of MLS operation (Oct. 1991 through Sep. 1992- left panels - and subsequent year - right panels -). From bottom to top, retrieval grid pressures are at 100 hPa, 46 hPa, and 22 hPa. Units are ppmv.

Note: Figure 2 & 3 may be combined into one and extended in time so as to cover until mid-March 1994, as Fig. 1. (+relabel dates)

The UARS Day is 1 on Sep. 12 1991, 112 on Jan. 12 1992, 478 on Jan. 1 1993. There are 10 "UARS months" (each with about 36 days) in each of the 2 years shown, with MLS data starting on 1 October.

FIG. 3. Same as Fig. 2, but for 10 hPa, 5 hPa (really 4.6 hPa), and 2 hPa (really 2.2 hPa).

FIG. 4. Equatorial ozone variation about the 2.5 year mean, as measured by MIS since shortly after launch. A 36 day smoothing (running average) has been applied to the data, to remove most of the artificial variation coupled to the UARS yaw cycle (see text). Panels give mixing ratio changes at various stratospheric pressures, from 100 hPa to 2 hPa.

FIG. 5. Top panel: integrated ozone column (in DU) from MLS daily data between 5S and 5N (area-weighted) versus time, for column down to 100 hPa (solid line), 46 hPa (dash-dot line), and 22 hPa (dashed line). Bottom panel: UKMO 12 GMT zonal mean winds (in m/s) for same time period, over the equator (solid line for 46 hPa, dashed line for 22 hPa). Positive wind is westerly.

FIG. 6. Time series of MLS zonal mean ozone column (in DU) above 100 hPa, for a time period identical to that of Fig. 1 (Oct 1991 through mid-March 1994) and for the same 16 degree increments in latitude. Three curves are shown for each year of data (same identification as in Fig. 1).

Vertical scales cover a range of 100 DU for all latitudes except for 80 and -80 degrees, where the variations over a year are larger, particularly in the south.

FIG. 7. Area-weighted average h41,S ozone column for various latitude bins (see labels), as a function of time since Oct 1991 (curves have same identification as in Figs. 1 and 6). Mean columns are computed above 100 hPa (top left panel), above 22 hPa (top right), between 100 and 46 hPa (bottom left), and between 46 and 22 hPa (bottom right).

FIG. 8. Column ozone (DU) from Nimbus-7 TOMS (solid line) and MLS for the 60 to 80 south latitude bin, for a time period covering the ozone hole development during the summer/spring of 1992. The various MIS data points are labeled with the pressure down to which the column is computed (TOMS measurements go down to the ground, in principle, which accounts for the larger values).

FIG. 9. Column ozone (DU) from TOMS and MLS measurements for the 30 to 60 degrees north latitude bin (top panel) and for 30 to 60 degrees south (bottom panel), during the Oct. 1991 through Sep. 1993 period. Nimbus-7 TOMS data are the solid lines (blue for 1991-92, red for the lower values of 1992-93). Meteor-3 TOMS data are shown by dotted green line, overlapping the Nimbus-7 TOMS results during the 1 Jan 1993 to 6 May 1993 interval. MLS column ozone above 100 hPa is shown as open blue circles for 1991-92, and as smaller filled red dots for 1992-93.

FIG. 10. MLS column ozone time series as a function of latitude for the period 1 Oct. 1991 through 30 Sep. 1992 (top panel), along with the following year (middle panel) and the difference - 2nd year minus the 1st - (bottom panel). Column values are computed above 100 hPa.

FIG. 11. Time evolution of the latitude/pressure cross section for ozone density (from MLS data, coupled with MLS and NMC temperatures), for roughly two week averages in the 1991-92 winter (top 4 panels), 1992-93 winter (middle 4 panels), as well as the difference - 2nd year minus the 1st - (bottom 4 panels). The exact 4 time periods (from left to right) are: 5 Dec to 21 Dec, 22 Dec to 8 Jan, 15 Feb to 28 Feb, and 1 Mar to 18 Mar. Note: (a) and (b) will be juxtaposed next to each other - (a) to the left of (b) - for final figure. Some further cosmetics are needed in this fig. (as well as some others).

FIG. 12. MLS and TOMS ozone column values for the 5 to 10 degrees south latitude bin, during the period 1 Oct. 1991 to 31 Dec. 1993. Top panel: Nimbus-7 TOMS total column data are represented as solid line, Meteor-3 data as dashed line (this overlaps the solid line for 1 Jan. 1993 to 6 May 1993), and MLS column (above 100 hPa) as dots. Bottom panel: Residual difference between the TOMS and MLS column amounts. Plus symbols are for Nimbus-7 TOMS minus MLS, and triangles are for Meteor-3 TOMS minus MLS.

FIG. 13. Residual column amount between TOMS and MLS data for the 5 to 10 degrees south latitude bin (same time period as in Fig. 12), for MLS column computed down to 100 hPa (solid line) and down to 46 hPa (dashed line). The lines are polynomial fits through the residual differences (the solid line corresponds to the data shown in the bottom panel of Fig. 12).

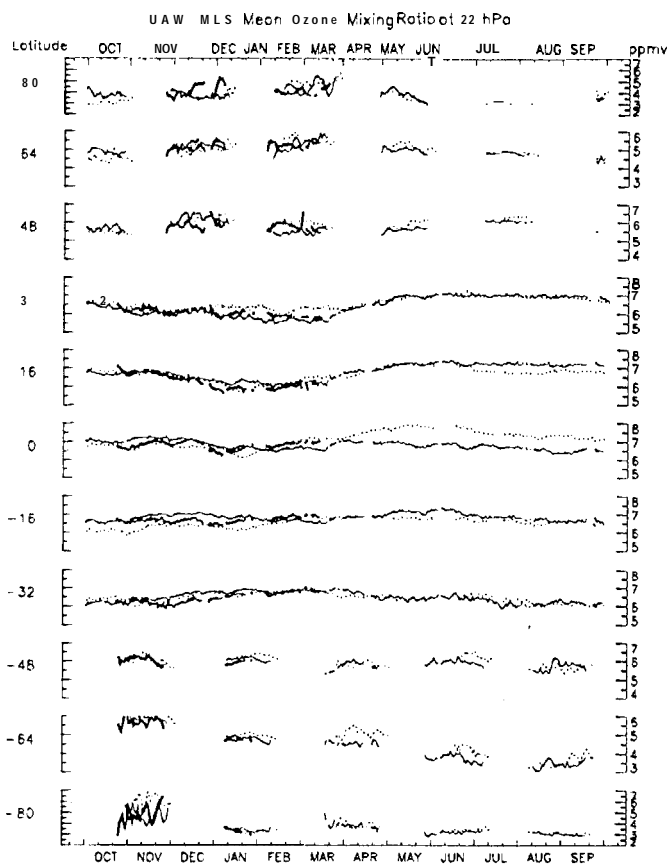
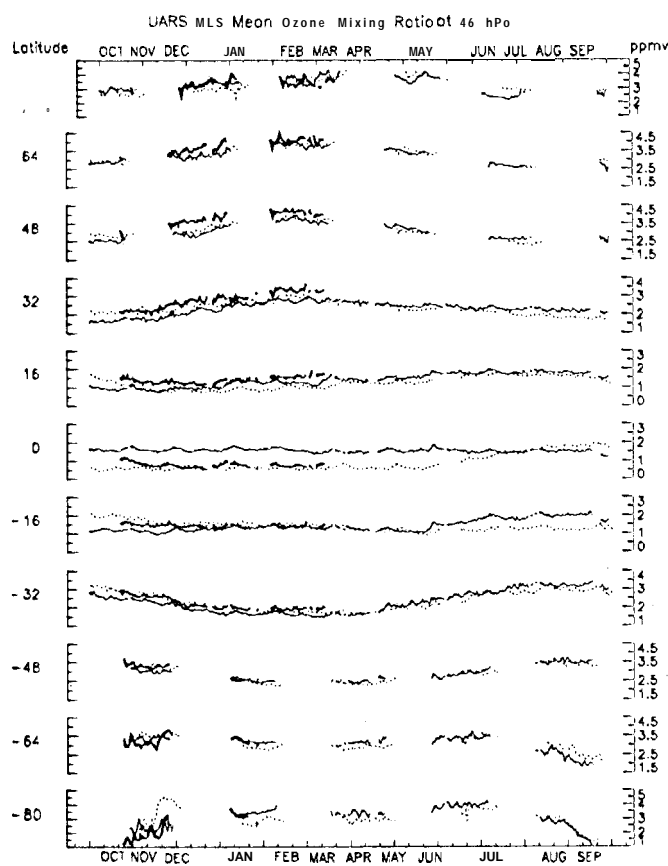
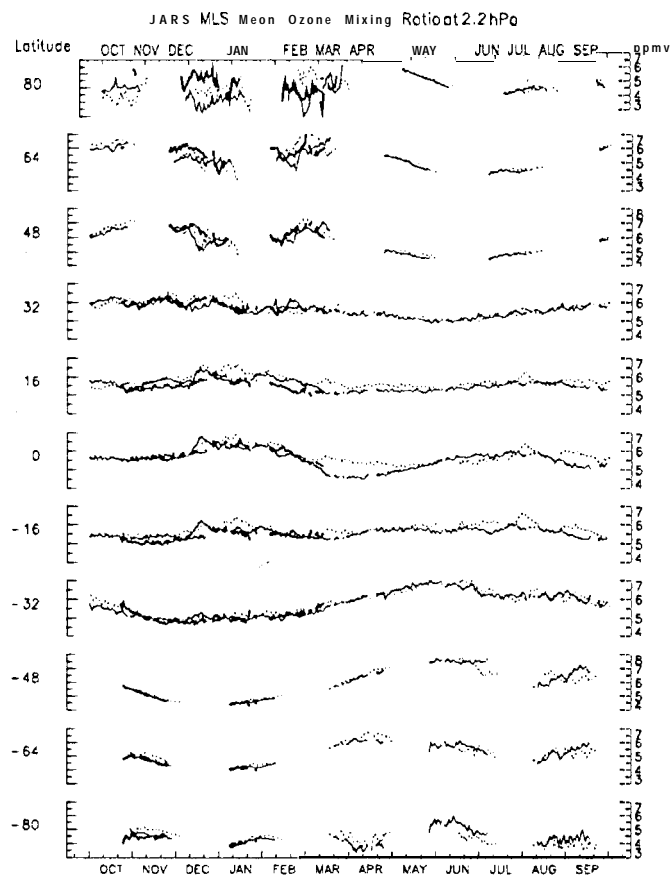
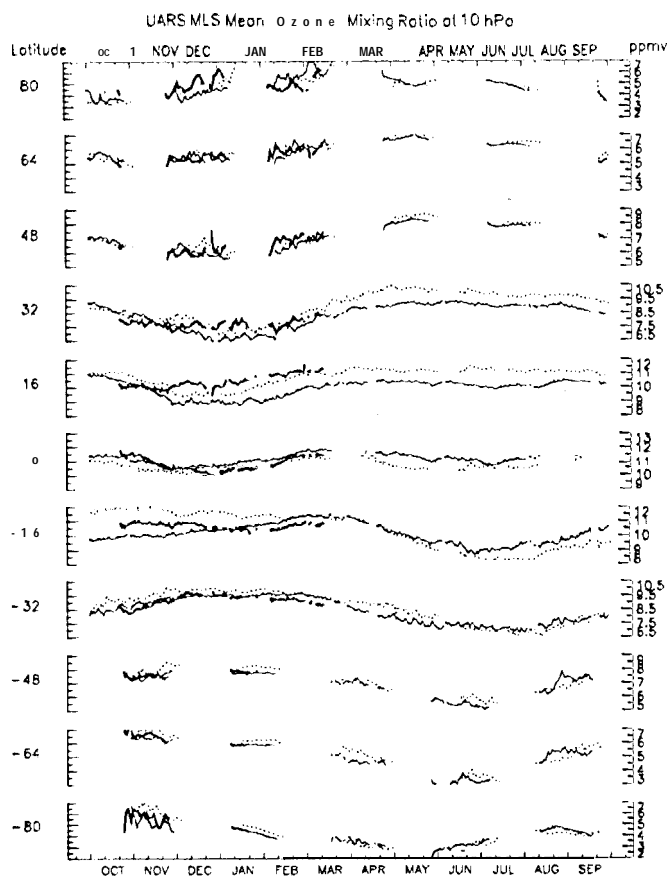


Fig. 1

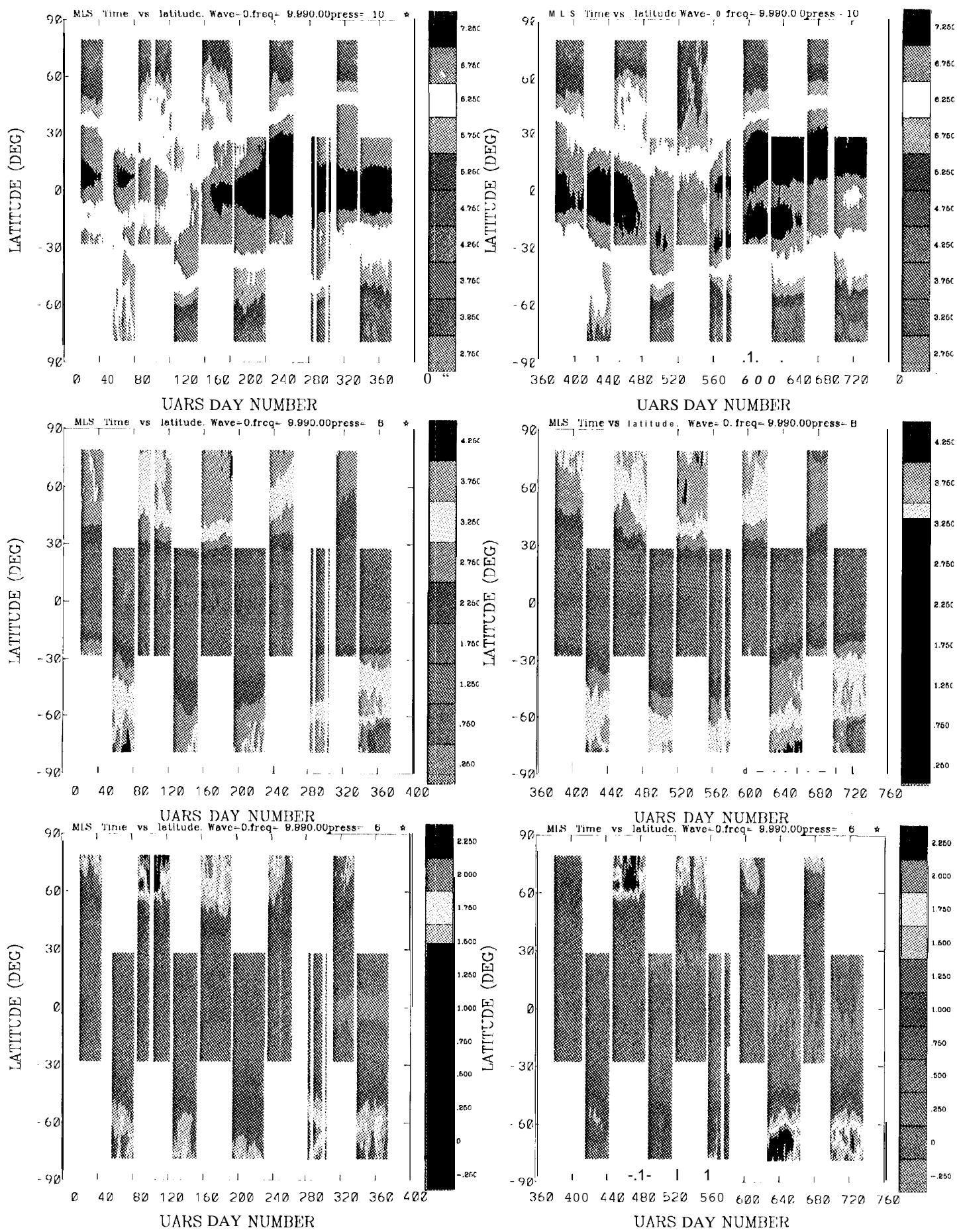


Fig. 2

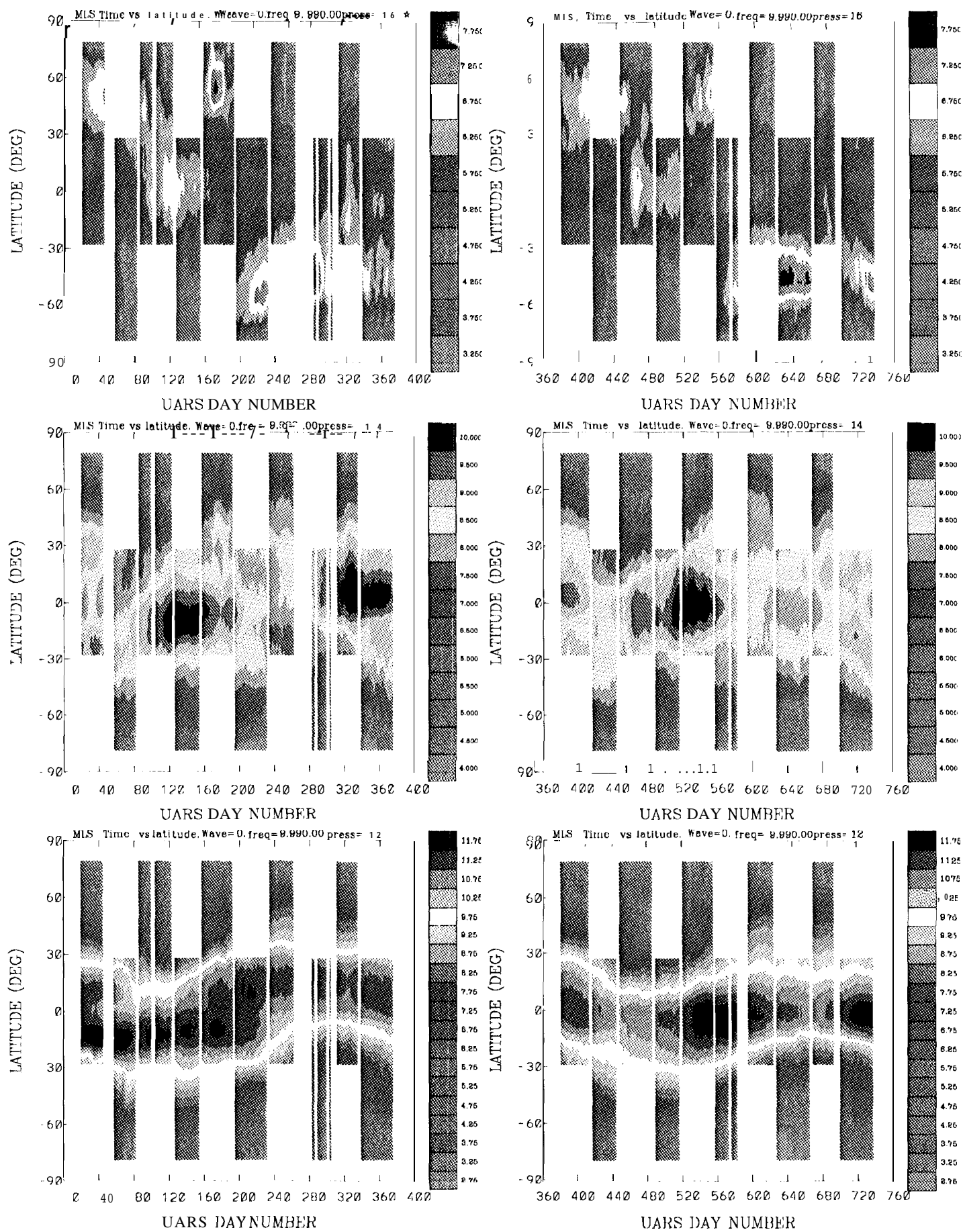


Fig. 3

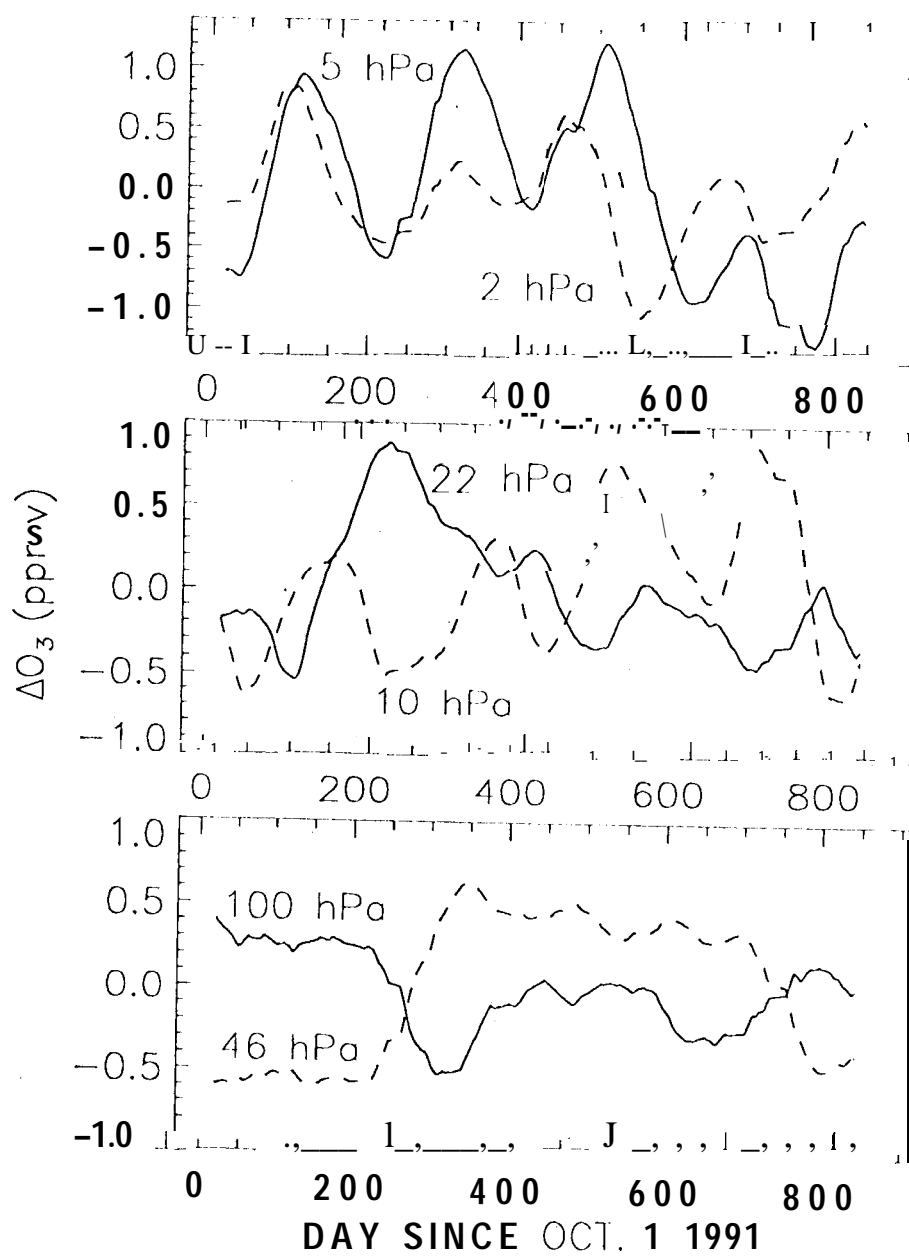


Fig. 4

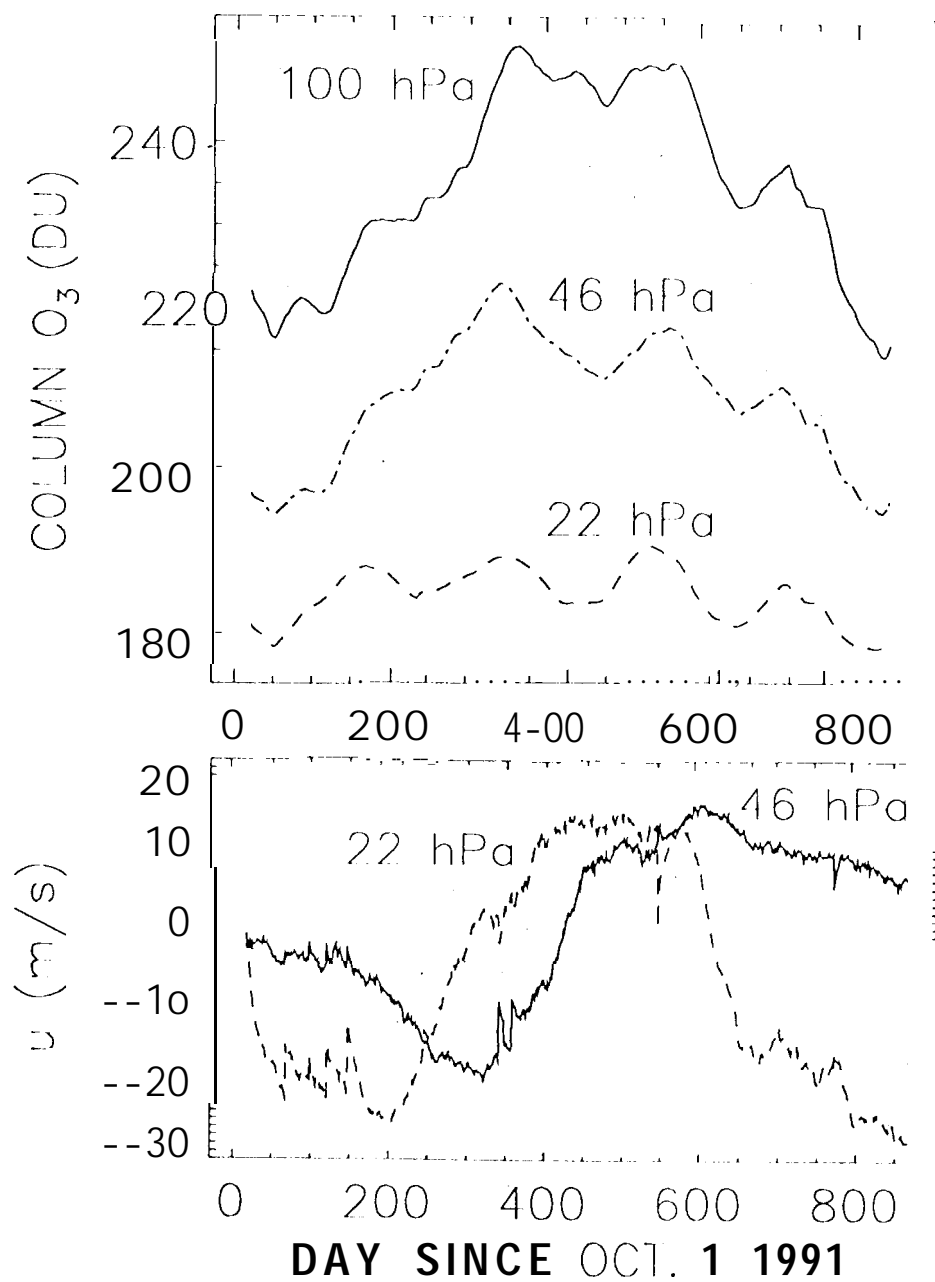


Fig. 5

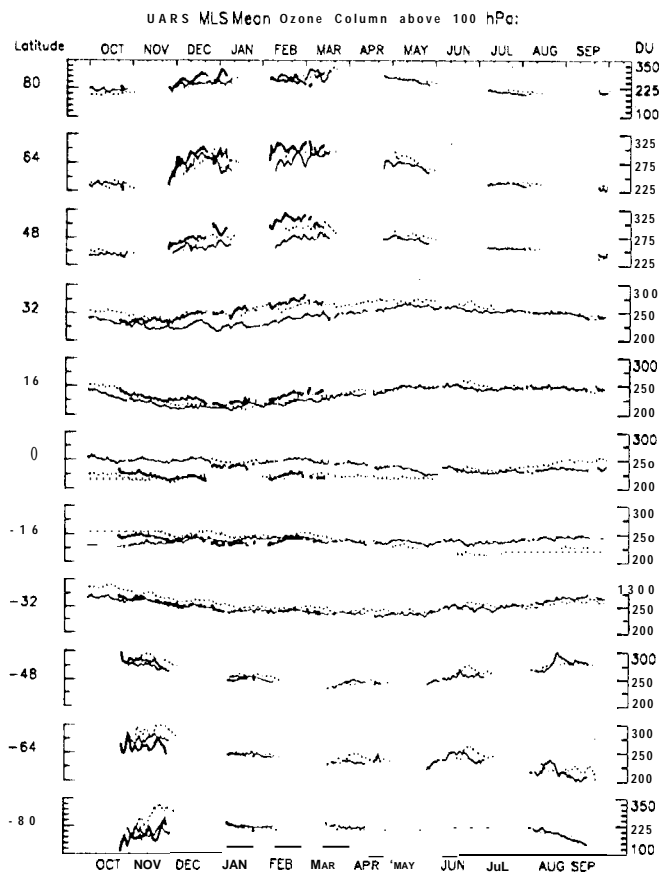


Fig. 6

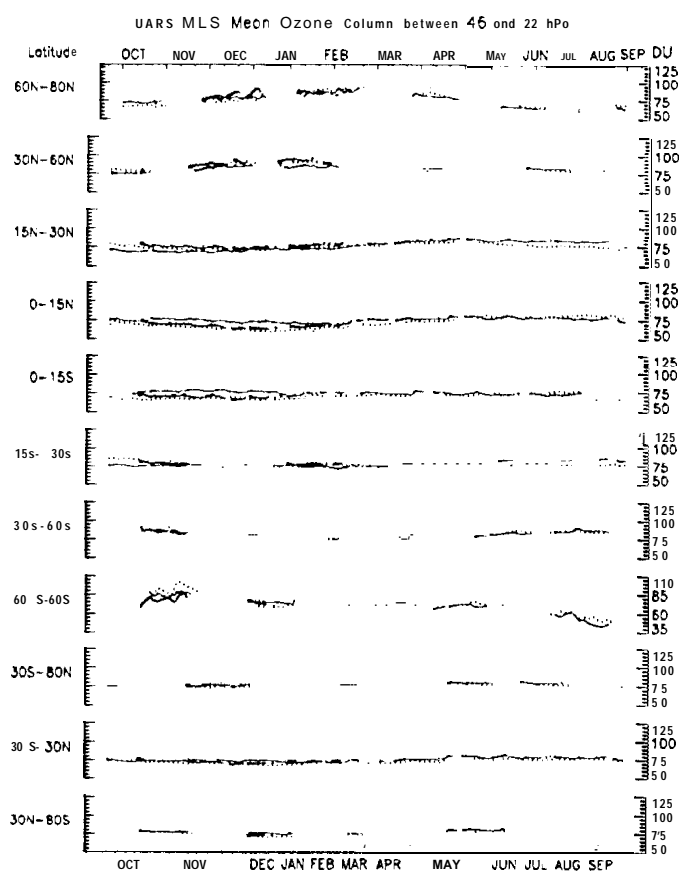
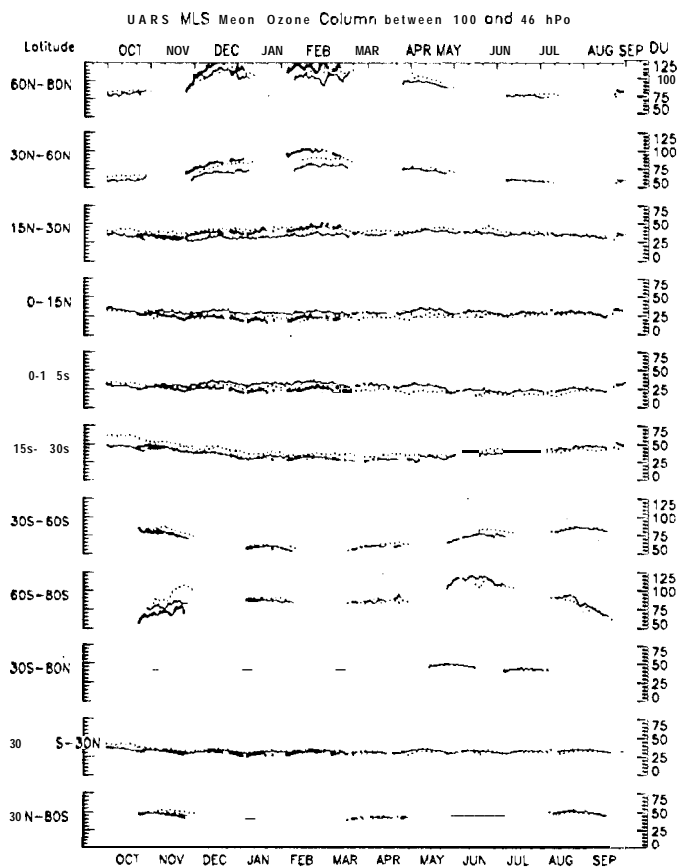
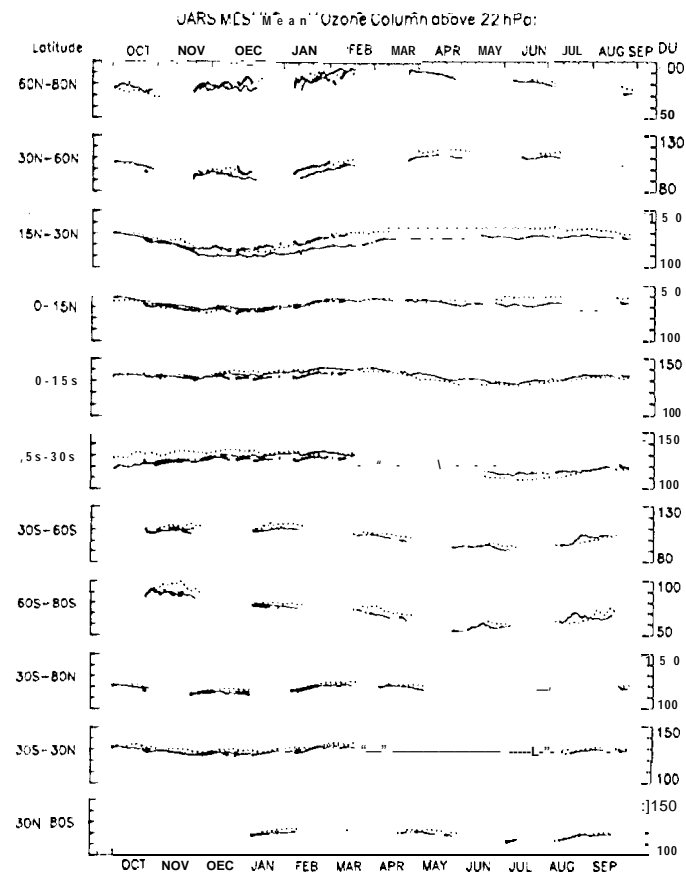
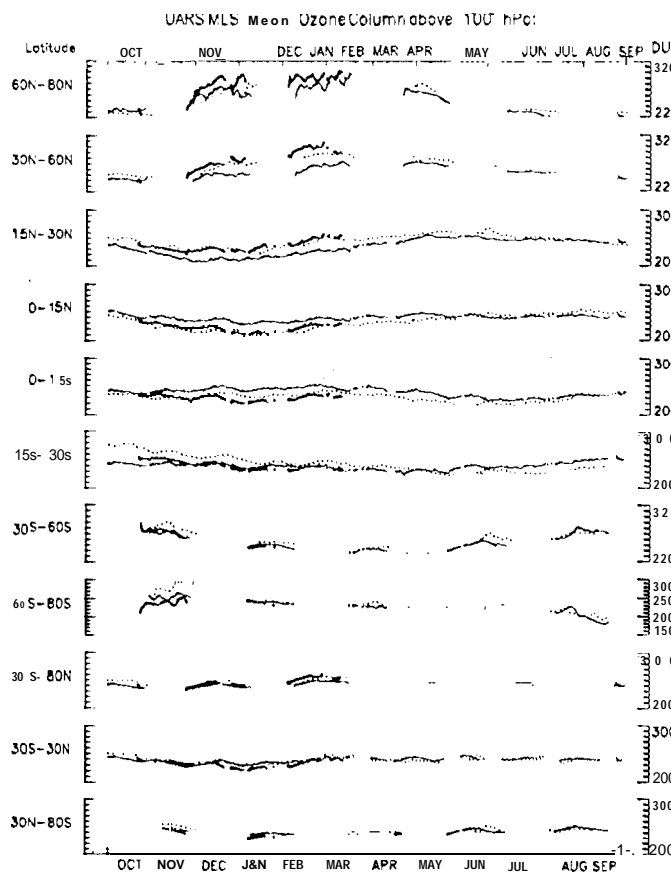


Fig-7

COL UMN O3 80S- 60S
UARS **MIS** AND NIMBUS 7 TOMS

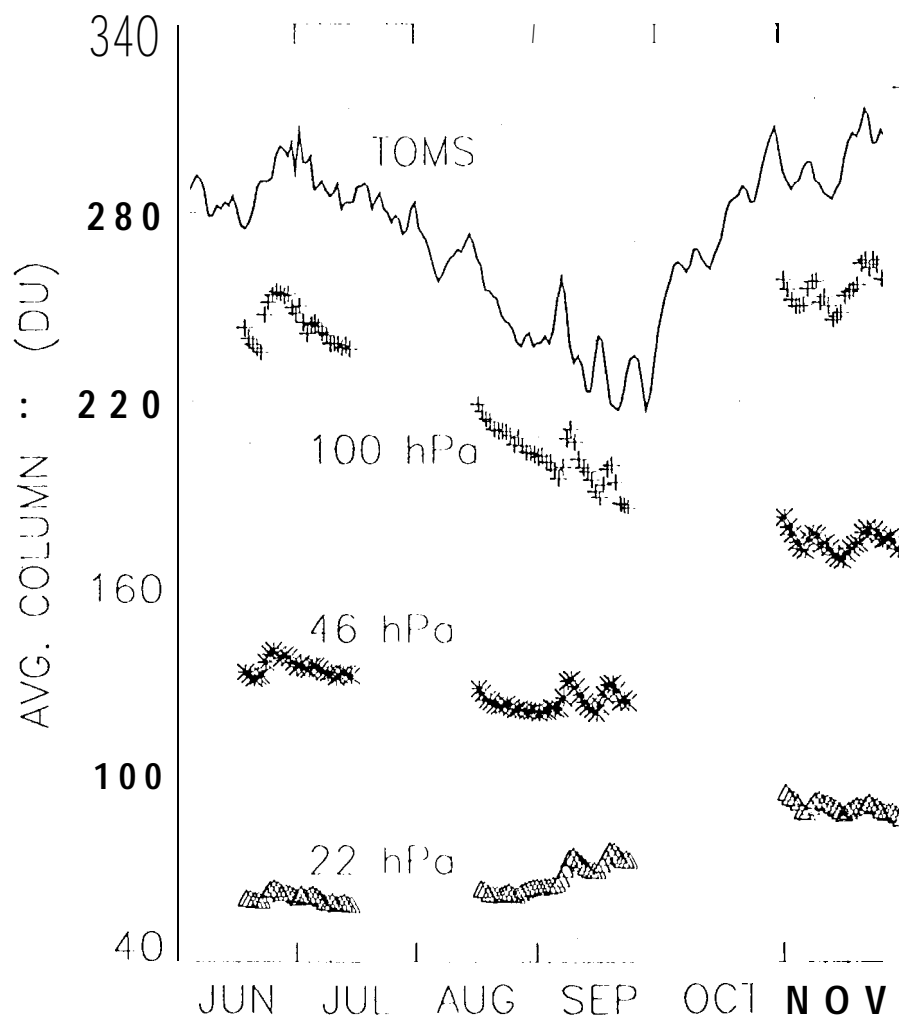
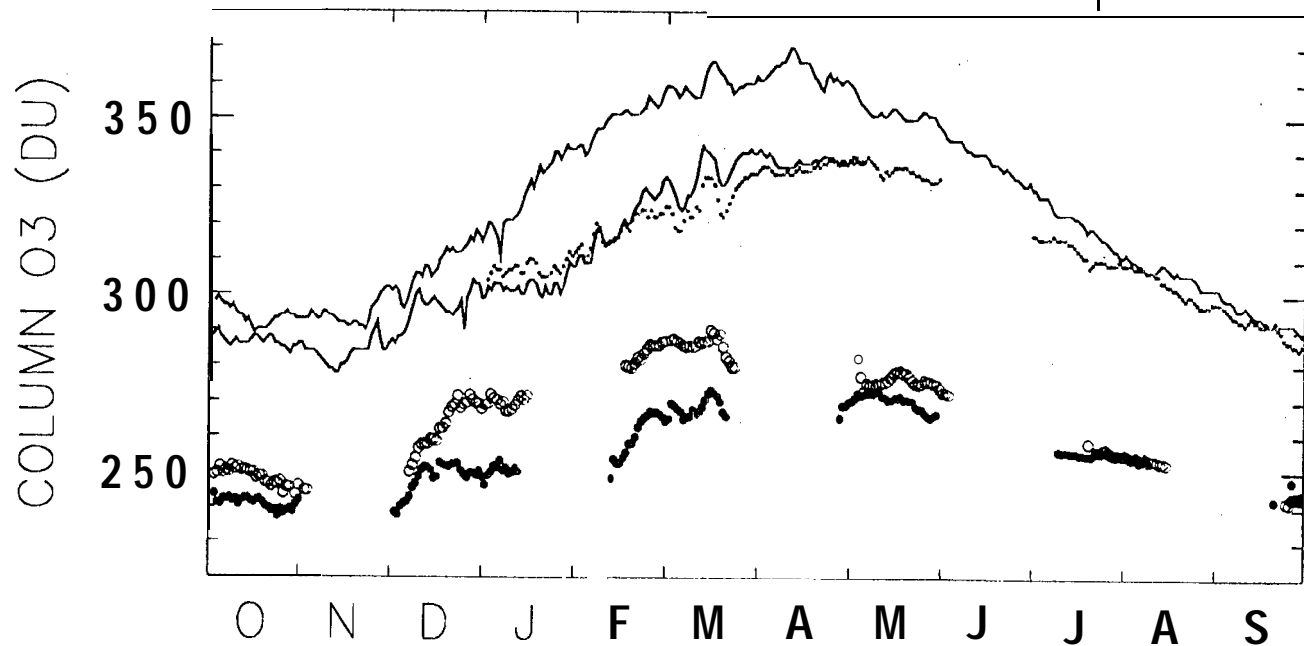


Fig. 8

AVG. COLUMN O3 30 N-60N
MLS & TOMS Oct. 91 - Sep. 93



AVG. COLUMN O3 60 S--30S
MLS & TOMS Oct. 91 - Sep. 93

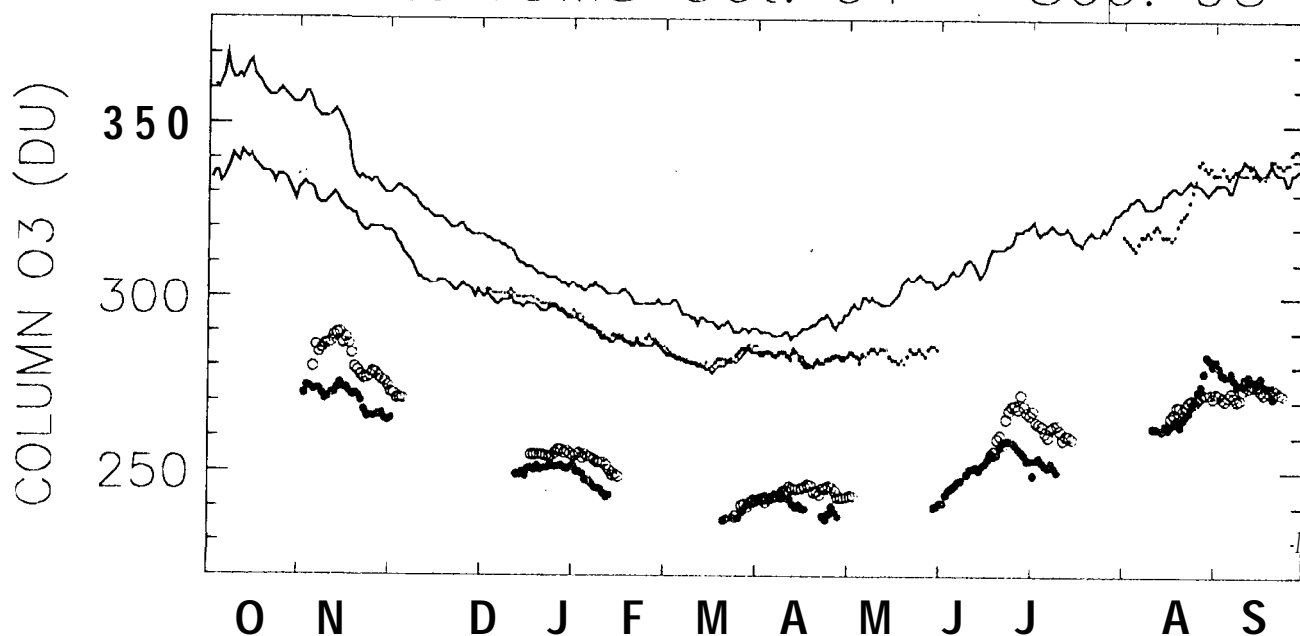
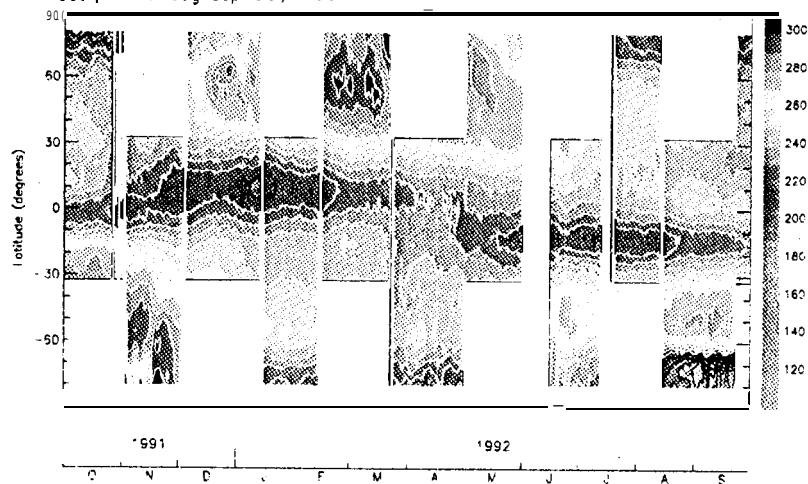


Fig. 9

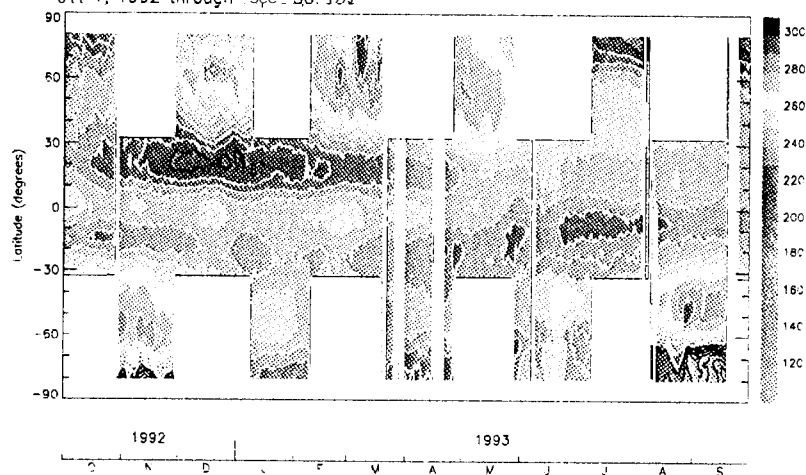
Column 03 (DU)
Above 100 hPa

Oct 1, 1991 through Sep 30, 1992



Column 03 (DU)
Above 100 hPa

Oct 1, 1992 through Sep 30, 1993



Column 03 (DU)
Above 100 hPa

Oct 1, 1992 through Sep 30, 1993 minus Oct 1, 1991 through Sep 30, 1992

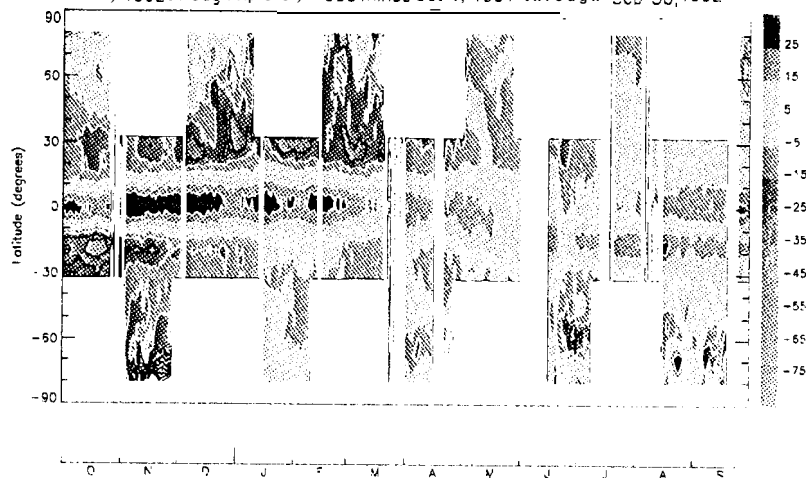
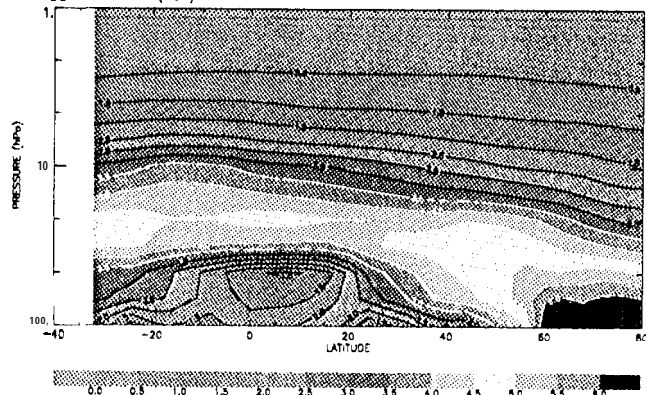


Fig. 10

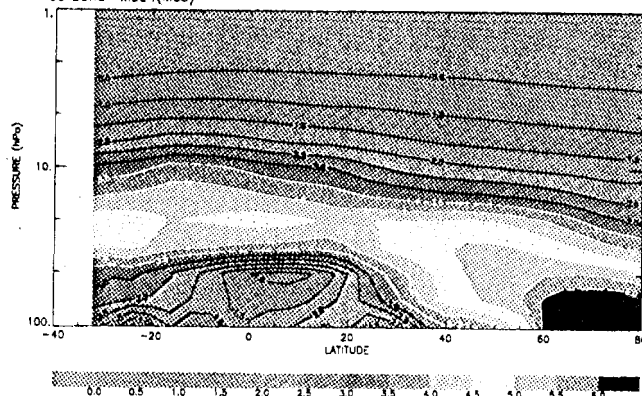
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 Limits: LAT: -90.0 to 90.0, LQ G: 0.0 to 360.0, SZA: 0.0 to 180.0, LST: 0.0 to 24.0
 03 Zonal Mean (MLS)



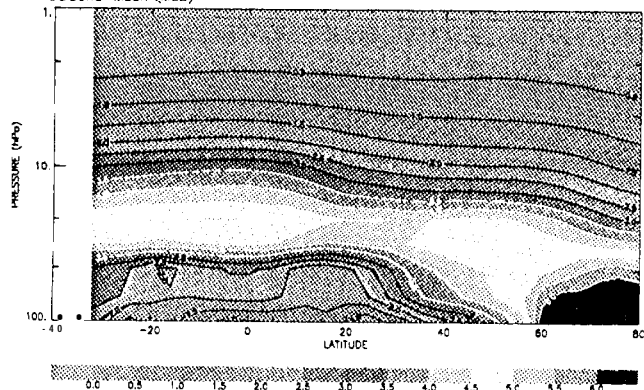
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 O3 DENSITY (1.e+12 mol/cm3)
 Limits: LAT: -90.0 to 90.0, LQ G: 0.0 to 360.0, SZA: 0.0 to 180.0, LST: 0.0 to 24.0
 03 Zonal Mean (MLS)



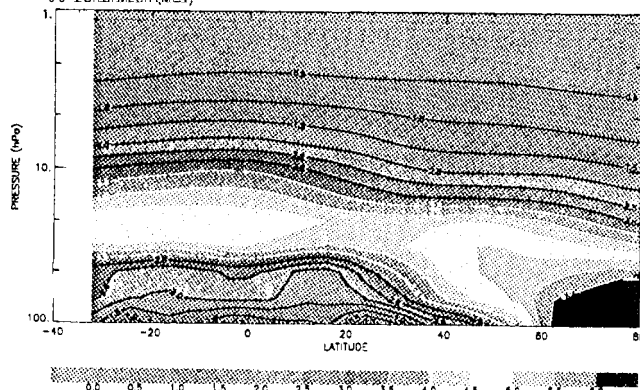
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 03 Zonal Mean (MLS)



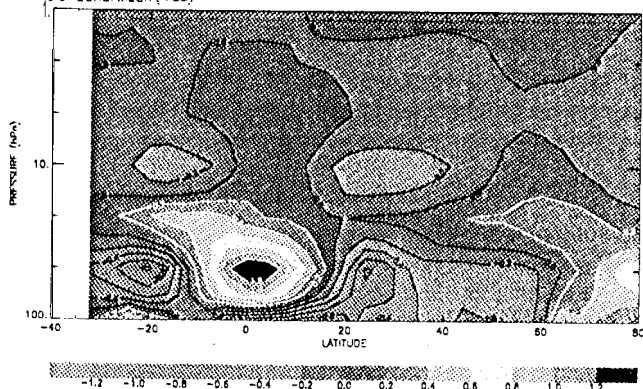
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 All data with MMAF stat = G
 O3 DENSITY (1.e+12 mol/cm3)
 Limits: LAT: -90.0 to 90.0, LQ G: 0.0 to 360.0, SZA: 0.0 to 180.0, LST: 0.0 to 24.0
 03 Zonal Mean (MLS)



LIARS day 451

/data/umis/13a/o3_205_d0451_v412.dot_1 + 16 days -
 /data/umis/13a/o3_205_d0085_v412.dot_1 + 16 days
 O3 DENSITY (1.e+12 mol/cm3)
 03 Zonal Mean (MLS)



UARS day 468

/data/umis/13a/o3_205_d0468_v412.dot_1 + 17 days -
 /data/umis/13a/o3_205_d0102_v412.dot_1 + 17 days
 O3 DENSITY (1.e+12 mol/cm3)
 03 Zonal Mean (MLS)

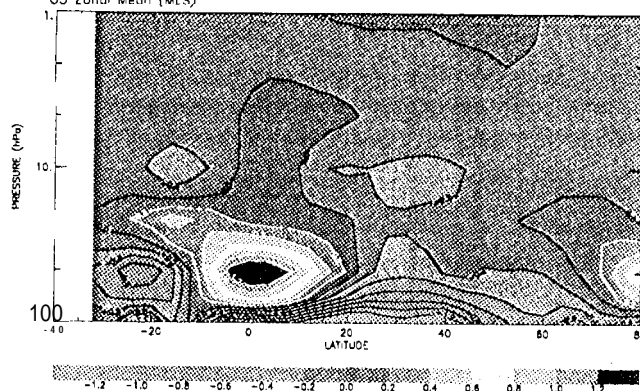


Fig. 11(a)

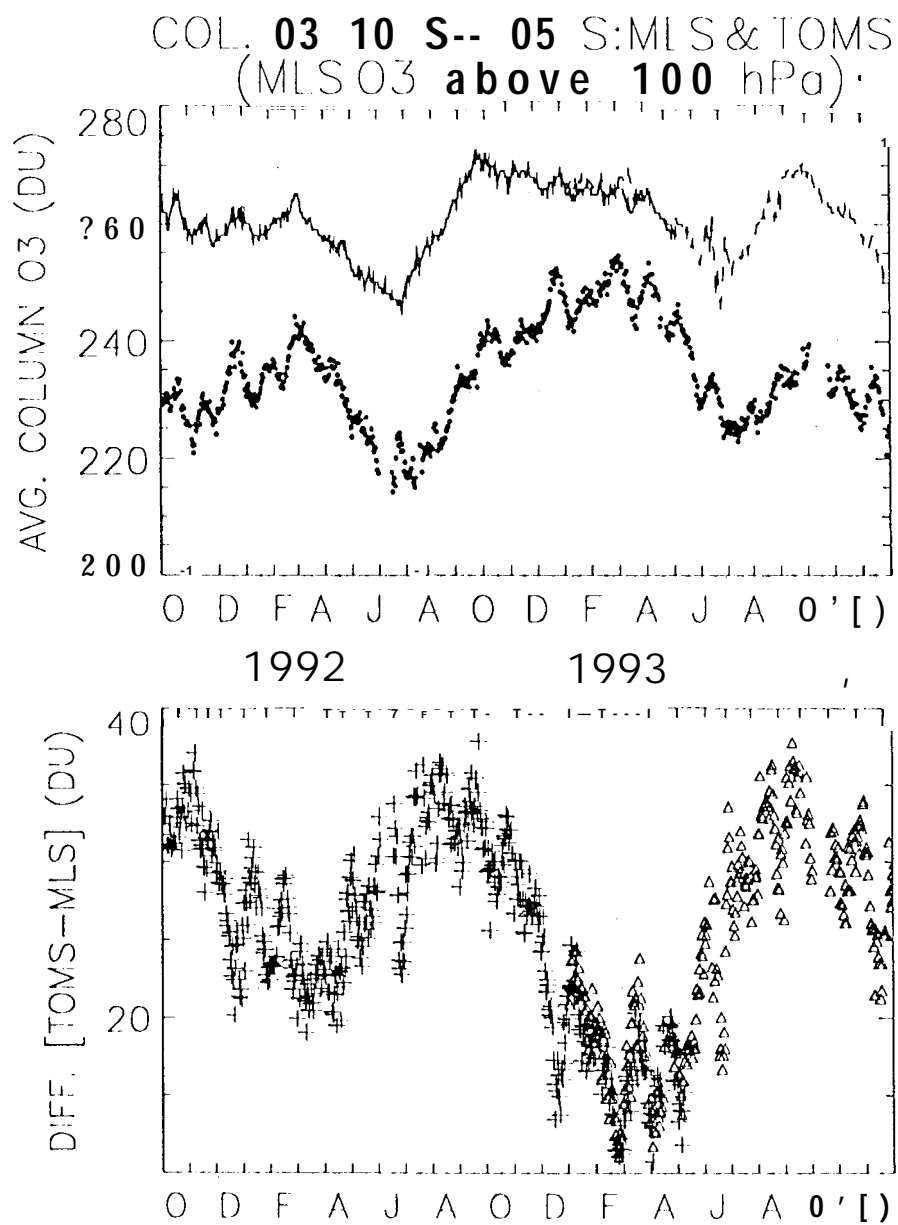


Fig. 12

RESIDUAL COLUMN OS 1 OS -- 05S

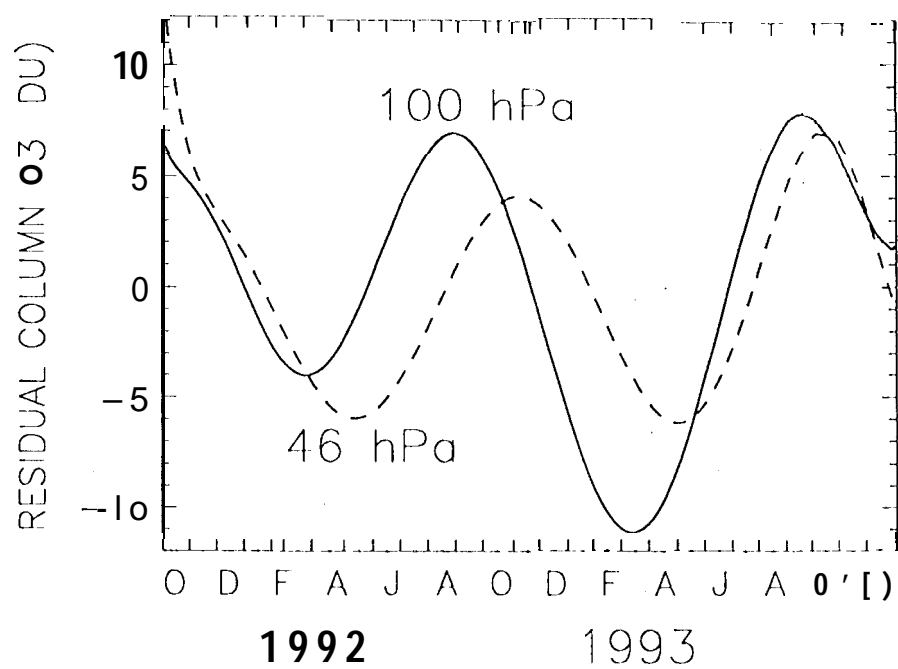


Fig. 13